United States
Environmental Protection
Agency

Office of Air Quality
Planning and Standards
Research Triangle Park, NC 27711

EPA-453/R-97-011
June 1997

Deposition of Air Pollutants to the Great Waters

Second Report to Congress

Printed on Recycled Paper

ACKNOWLEDGEMENTS

This report was prepared by the U.S. Environmental Protection Agency's (EPA's) Office of Air Quality Planning and Standards (OAQPS).

EPA wishes to thank the authors for the time and effort they spent preparing and reviewing the drafts of the document. The lead contributors to the writing, editing, and review of this document were members of the OAQPS's Great Waters Team: John Ackermann, Melissa McCullough, Eric Ginsburg, Dianne Byrne, and Laurel Driver. Recognizing the breadth of the task of preparing this report, OAQPS worked with individuals in EPA offices to coordinate input into the report. These individuals represent the "Great Waters Core Group" and include the following individuals: Angela Bandemehr, Rich Batiuk, Gary Evans, Jim Giattina, Martha Keating, Maggie Kerchner, Virginia Kibler, Carl Nash, Doris Price, Roy Smith, and Joe Touma.

In addition, substantial review of the document also was provided by the following EPA offices:

Chesapeake Bay Program Office
Gulf of Mexico Program Office
Great Lakes National Program Office
National Health and Environmental Effects Research Laboratory
Office of Air and Radiation
Office of Policy, Planning and Evaluation
Office of Research and Development
Office of Water
Region II
Region V

The following organizations also have been included in review of the document:

Atmospheric Environment Service, Environment Canada
Centre for Atmospheric Research Experiments, Environment Canada
Department of Commerce/National Oceanic and Atmospheric Administration (NOAA)
Environmental Defense Fund
Maine Department of Environmental Protection
Michigan Department of Environmental Quality
Wisconsin Department of Natural Resources
Wisconsin Electric Power Company and Electric Power Research Institute

In addition, this document was reviewed by a panel of external scientific experts whose constructive comments provided critical assistance in improving the scientific perspective of the final report. These experts include:

Dr. Anders W. Andren	Sea Grant Institute, University of Wisconsin-Madison
Dr. William F. Fitzgerald	Department of Marine Sciences, University of Connecticut
Dr. Louis J. Guillette Jr.	Department of Zoology, University of Florida
Dr. Hans W. Paerl	Institute of Marine Sciences, University of North Carolina

The following individuals also provided specific comments on the draft document: Mary Ann Allan, Richard Artz, Terry F. Bidleman, Rona Birnbaum, Marty Burkholder, David Cleverly, Ellen Cooter, Carolyn Currin, Stan Durkee, David W. Evans, Theresa Faber, Peter

Finkelstein, Jim Galloway, Ellen Heath, Steven F. Hedtke, Roland Hemmett, Bruce Hicks, Raymond M. Hoff, Suzanne King, Peter Landrum, Nancy Laurson, Suzanne McMaster, David T. Michaud, Todd Nettesheim, Ellen E. Parr Doering, Jon Taylor, Mike Thrift, Rick Tonielli, and Richard Valigura.

Technical support in preparing this document has been provided by ICF Kaiser under EPA Contract Nos. 68D4-0103 and 68D6-0064. The efforts of Karen Gan and Anne Cowan are especially appreciated.

Appreciation is given to all others who have contributed to this report and were inadvertently not mentioned above.

EXECUTIVE SUMMARY

This report is the Second Report to Congress on the atmospheric deposition of pollutants to the Great Waters. Section 112 of the Clean Air Act (CAA) provides the legislative basis for hazardous air pollutant (HAP) programs directed by the U.S. Environmental Protection Agency (EPA). In response to mounting evidence that air pollution contributes to water pollution, Congress included section 112(m), *Atmospheric Deposition to Great Lakes and Coastal Waters*, in the 1990 CAA. Under this statute, EPA is required to periodically report to Congress on the results of this program. Concurrent with the Second Report to Congress, EPA is to determine the adequacy of section 112 to prevent adverse effects to public health and serious or widespread environmental effects associated with atmospheric deposition of HAPs to the Great Waters.

How does this report differ from the 1994 Report to Congress?

The First Report to Congress presented information about the health and environmental effects associated with the pollutants of concern, relative atmospheric loadings, and the potential sources of these loadings. The current report documents findings since the First Report to Congress and describes recent progress in these issues. This report places emphasis on local and federal activities, including many that support section 112(m) directives, taking place at specific waterbodies such as the Great Lakes, Lake Champlain, and Chesapeake Bay, as well as coastal estuaries designated through the National Estuary Program and National Estuarine Research Reserve System. Due to the short time period since the First Report to Congress, much of the research data collected during this time are still in the process of being analyzed; however, the objectives and status of these efforts are described in the report. Furthermore, this report does not assess the linkage between the potential sources, loadings, and effects of pollutants of concern because, as in the First Report to Congress, the scientific information is currently not sufficiently complete. As such, unanswered questions still remain as well as uncertainties for some issues. This report proposes a number of future directions to reduce uncertainty in several areas.

Because this report is an update of the First Report to Congress, the information presented here cannot be used alone to develop recommendations regarding atmospheric deposition of pollutants to the Great Waters. Rather, the scientific information summarized in this report, together with the findings and recommendation identified in the First Report to Congress, can be used to assess the extent of progress as a result of recommendations from the First Report to Congress and to determine what gaps in information still exist.

Has the list of Great Waters pollutants of concern changed?

The pollutants of concern to the Great Waters have not changed since the First Report to Congress. The list consists of 15 pollutants (see sidebar on next page) including pesticides, metal compounds, chlorinated organic compounds, and nitrogen compounds. These pollutants have been selected based on information regarding their health and environmental effects and evidence that they are atmospherically deposited to the Great Waters. Most are bioaccumulative chemicals that persist in the environment for long periods. Many of these pollutants are listed as chemicals of concern on toxics lists for individual waterbodies at the local and statewide level.

What are the environmental and public health effects of the pollutants of concern to the Great Waters?

The 15 Great Waters Pollutants of Concern

- Cadmium and cadmium compounds
- Chlordane
- DDT/DDE
- Dieldrin
- Hexachlorobenzene (HCB)
- α-Hexachlorocyclohexane (α-HCH)
- Lindane (γ-hexachlorocyclohexane; γ-HCH)
- Lead and lead compounds
- Mercury and mercury compounds
- Polychlorinated biphenyls (PCBs)
- Polycyclic organic matter (POM)
- Tetrachlorodibenzo-p-dioxin (TCDD; dioxins)
- Tetrachlorodibenzofuran (TCDF; furans)
- Toxaphene
- Nitrogen compounds

Recent scientific information confirms adverse effects data presented in the First Report. The pollutants are associated with deleterious effects on many target organs in humans and animals, including the liver, kidney, nervous system, endocrine system, reproductive organs, and immunological system. Few new developments have occurred in this area, although there is a growing interest about the potential for some pollutants to act on and disrupt the endocrine system in wildlife and possibly in humans.

As in the First Report to Congress, the contribution of atmospheric deposition of the pollutants and subsequent exposure to potential human health and ecological effects cannot be quantified at this time. Pollutants deposited from the air directly into a waterbody may have routes of exposure to aquatic life that differ from exposure by waterborne inputs; however, there are few studies available to address this issue. There is currently no information to suggest that effects produced by pollutants deposited from the air will be different from effects by these pollutants carried in water or found in sediment. Contamination in fish can enter the diet of humans and other animals and, therefore, fish-eating birds or mammals are especially at risk from pollutants that biomagnify because they are exposed to concentrated levels of these pollutants. Evaluation of potential human health effects of pollutants of concern is based almost completely on laboratory studies in animals. The data from these studies may be extrapolated to assess potential adverse effects in humans; however, uncertainties may exist as to the exposure levels at which these potential effects may occur. Atmospheric deposition of nitrogen compounds can contribute significantly to eutrophication in coastal waters, where plant productivity is usually limited by nitrogen availability. Accelerated eutrophication and its subsequent effects such as nuisance algal blooms and reduced oxygen levels pose significant problems for Chesapeake Bay and many other estuaries.

Do water quality exceedances or fish advisories continue to occur as a result of pollution loadings to the Great Waters?

Current water quality criteria exceedances and fish advisories suggest that toxic contamination by persistent toxics is present in the Great Waters. The contribution of atmospheric deposition to the water quality exceedances and contaminant levels in fish is not known at this time. More information on relative loadings of pollutants is needed to assess the extent of contamination attributed to atmosphere.

Water quality criteria have been developed specifically for the Great Lakes, and exceedances of these criteria continue to occur. Recent information is available for some pollutants, and in general, these exceedances have declined in recent years. Fish advisories that are issued by states for individual pollutants provide qualitative information about potential exposure and the extent of contamination in a waterbody. PCBs are most commonly the focus of fish advisories issued in the Great Waters and their basins, with dioxins having the next highest occurrence of advisories. Elevated levels in fish of other pollutants, such as chlordane and mercury, also have warranted fish advisories in many states around the Great Waters.

What is currently known about the atmospheric deposition of the pollutants of concern to the Great Waters?

The contribution of atmospheric deposition to overall pollutant loadings in the Great Waters continues to be studied. Atmospheric loadings of pollutants result from wet deposition and dry particle deposition and through air-water gas exchange. Described in this report are monitoring and modeling studies relevant to atmospheric deposition that are currently taking place at the major waterbodies of the Great Waters.

Recent atmospheric monitoring data from a binational monitoring network assessing trends of atmospheric deposition to the Great Lakes region indicate that atmospheric levels of toxic pollutants are declining slightly or leveling off and remain a significant concern in the Great Lakes. Several recent activities in the Great Lakes have been initiated to characterize and reduce toxic contamination and deposition to these waters. In the Lake Champlain basin, research on atmospheric loading of mercury is currently underway in the basin. Early data show that atmospheric mercury levels and deposition are comparable to those measured around the Great Lakes.

Nitrogen and toxic contaminants are a concern in Chesapeake Bay and other coastal waters. Excessive nitrogen loading can accelerate eutrophication and its adverse effects, such as nuisance algal blooms and fish kills. Substantial progress has been made in addressing nitrogen contamination issues in Chesapeake Bay, the largest U.S. estuary. A strategy has been developed by the Chesapeake Bay Program for reducing the nitrogen load to the Bay. Part of this process includes the large-scale modeling and understanding of the type and geographic origin of airborne nitrogen to the Bay. Significant data also have been collected on rates and amounts of nitrogen deposition (including comparison of direct and indirect deposition and of wet and dry deposition), and models have been developed to evaluate the impact of several nitrogen reduction scenarios on the Bay's water quality.

Since the First Report to Congress, studies of other coastal waters, at National Estuary Program waters in particular, have investigated the significance of atmospheric deposition of nitrogen compounds to their waters. To improve understanding and reduction of nitrogen deposition to Chesapeake Bay and other coastal waters, the Chesapeake Bay Program, various National Estuary Programs, and the Gulf of Mexico Program continue to develop and refine modeling and monitoring efforts by addressing uncertainties such as nitrogen retention in watersheds, the differences in transport and fate of various nitrogen compounds, and the contribution of nearshore ocean waters to the nitrogen inputs to estuaries.

What is currently known about the sources of atmospheric pollutant deposition to the Great Waters?

Both local and long-range emission sources contribute to atmospheric deposition in the Great Waters. Emission inventories on specific sources of the pollutants of concern are actively being developed and efforts to gather more information on the potential sources of contamination continue. For example, EPA recently completed a national emissions inventory of known U.S. sources of seven hazardous pollutants of concern listed under CAA section 112(c)(6). Identification of the sources for total emissions of these pollutants is leading to an evaluation of the stationary, anthropogenic source categories to determine whether they are currently regulated or scheduled for regulation under the CAA. Some persistent pollutants are no longer produced through human activities but may continue to affect the Great Waters environment through releases from existing equipment and repeated cycling between the atmosphere, land, and waterbodies.

Understanding atmospheric processes is necessary for analyzing the relationships between source emissions, relative loadings, and the potential for adverse effects in humans and the environment. Because it is often difficult to establish these relationships clearly and quantitatively through available measurement data (e.g., it can be difficult to differentiate between the contribution of distant versus local sources to the loading of a pollutant to a particular waterbody), investigators frequently use mathematical models of atmospheric transport and deposition. This report presents the application of several atmospheric transport and deposition models to the Great Waters and how these models compared to actual data from the waterbodies. Extensive modeling of nitrate emissions and transport that can deposit to Chesapeake Bay has calculated the "airshed" of distant as well as local sources.

What are EPA's current conclusions from this Second Report to Congress?

The information presented in this report advances scientific knowledge on issues related to atmospheric deposition of pollutants to the Great Waters and confirms the findings and conclusions presented in the First Report to Congress. In general, concentrations of some persistent pollutants in the Great Lakes, as monitored by sample measurements of contaminant levels in the air, water, and biota, appear to have leveled off or declined only slightly in recent years.

EPA also has issued draft determinations that the provisions of CAA section 112 are adequate to prevent serious adverse human health effects and serious or widespread environmental effects as a result of atmospheric deposition of HAPs emitted by domestic stationary sources. At this time, EPA believes that there is no information to suggest that additional regulations beyond those authorized or required by section 112 are necessary or appropriate to prevent such effects. The draft determinations will be issued for public notice and comment by June 30, 1997, and final determinations will be made by March 15, 1998.

What future directions may be taken by EPA to support section 112(m)?

Described throughout the report are activities that have increased our knowledge of atmospheric deposition of pollutants to the Great Waters. As new information becomes available on atmospheric pollutant deposition to the Great Waters, additional questions or issues are expected to arise that will require further investigation or action. At this time, EPA has identified the following areas where information is limited and some specific steps that need to be taken to advance our understanding of issues relevant to the Great Waters program:

- Define and proceed with management and regulatory actions for Great Waters pollutants of concern, with a particular focus given to pollutants currently being emitted to the air from sources that can be subject to regulations under the CAA (for example, the seven pollutants of concern in section 112(c)(6));

- Continue to support monitoring and research efforts on deposition to make informed management decisions and to track reductions;

- Perform exposure and effects studies that will build on the recent Great Lakes Water Quality Criteria, which consider biomagnification. These studies will be coordinated with an integrated research strategy on the persistent pollutants, their distribution and concentrations, exposure routes, and associated effects;

- Improve modeling efforts to estimate atmospheric loadings to Great Waters. For example, adapt and apply the comprehensive approach developed for the Lake Michigan Mass Balance Model to additional waterbodies;

- Increase efforts to identify specific emissions sources of atmospheric deposition to the Great Waters, both nearby and relatively distant from the waterbody, to develop risk management strategies, as well as investigate the impact from cycling of pollutants that are no longer used or manufactured in the United States;

- Continue to promote pollution reduction in the Great Waters through local, regional, and federal initiatives, as well as coordinated international efforts; and

- Assess economic costs and benefits associated with reductions of pollutants to the Great Waters, including identifying and quantifying, where possible, economic impacts associated with exposure and effects indicators such as fish advisories, habitat decline, diminished species diversity, fish kills, and declining or contaminated shellfish and fish populations.

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Ah	Aryl hydrocarbon
AOC	Area of Concern
AQSM	Air quality simulation model
ATSDR	Agency for Toxic Substances and Disease Registry
AWQC	Ambient water quality criterion or criteria
B(a)P	Benzo(a)pyrene
CAA	Clean Air Act
CBADS	Chesapeake Bay Atmospheric Deposition Study
CBOS	Chesapeake Bay Observing System
CBP	Chesapeake Bay Program
CCMP	Comprehensive Conservation and Management Plan
CMB	Chemical mass balance
CWA	Clean Water Act
DDD	Dichlorodiphenyldichloroethane
DDE	Dichlorodiphenyldichloroethylene
DDT	Dichlorodiphenyltrichloroethane
DES	Diethylstilbestrol
dL	Deciliter
EMAP	Environmental Monitoring and Assessment Program
EPA	U.S. Environmental Protection Agency
FDA	Food and Drug Administration
g	Gram
GLWQA	Great Lakes Water Quality Agreement
GLWQB	Great Lakes Water Quality Board
GLWQC	Great Lakes Water Quality Criteria
GLWQG	Great Lakes Water Quality Guidance
GLWQO	Great Lakes Water Quality Objective
GMP	Gulf of Mexico Program
HAP	Hazardous air pollutant
HCB	Hexachlorobenzene
α-HCH, γ-HCH	alpha-Hexachlorocyclohexane, gamma-Hexachlorocyclohexane
Hg	Mercury
IADN	Integrated Atmospheric Deposition Network
IJC	International Joint Commission
kg	Kilogram
km, km^2, km^3	Kilometer, square kilometer, cubic kilometer
L	Liter
LaMP	Lakewide Management Plan
LCBP	Lake Champlain Basin Program
LMUATS	Lake Michigan urban air toxics study
LQER	Lesser-quantity emission rates
MACT	Maximum achievable control technology
MCL	Maximum contaminant level
m^2, m^3	Square meter, cubic meter
μg, ug	Microgram
mg	Milligram
NAAQS	National ambient air quality standard
NADP	National Atmospheric Deposition Program
NAPAP	National Acid Precipitation Assessment Program
NEP	National Estuary Program
NERRS	National Estuarine Research Reserve System
ng	Nanogram

NOAA	National Oceanic and Atmospheric Administration
NO_x	Oxides of nitrogen
NS&T	National Status and Trends
OAQPS	Office of Air Quality Planning and Standards
OTC	Ozone Transport Commission
PAH	Polycyclic aromatic hydrocarbon
PCA	Principal component analysis
PCB	Polychlorinated biphenyl
pGLWQC	Proposed Great Lakes water quality criteria
POM	Polycyclic organic matter
ppb, ppm	Parts per billion, parts per million
RADM	Regional Acid Deposition Model
RAP	Remedial Action Plan
RAPIDS	Regional Air Pollutant Inventory Development System
RELMAP	Regional Lagrangian Model of Air Pollution
REMSAD	Regional Modeling System for Aerosols and Atmospheric Deposition
RPM	Regional Particulate Model
SAB	Science Advisory Board
SAV	Submerged aquatic vegetation
SETAC	Society of Environmental Toxicology and Chemistry
SOLEC	State of the Lakes Ecosystem Conference
TBADS	Tampa Bay Atmospheric Deposition Study
TCDD	Tetrachlorodibenzo-p-dioxin
TCDF	Tetrachlorodibenzofuran
TRIADS	Texas Integrated Atmospheric Deposition Study
TSCA	Toxic Substances Control Act
VOC	Volatile organic compound
yr	Year

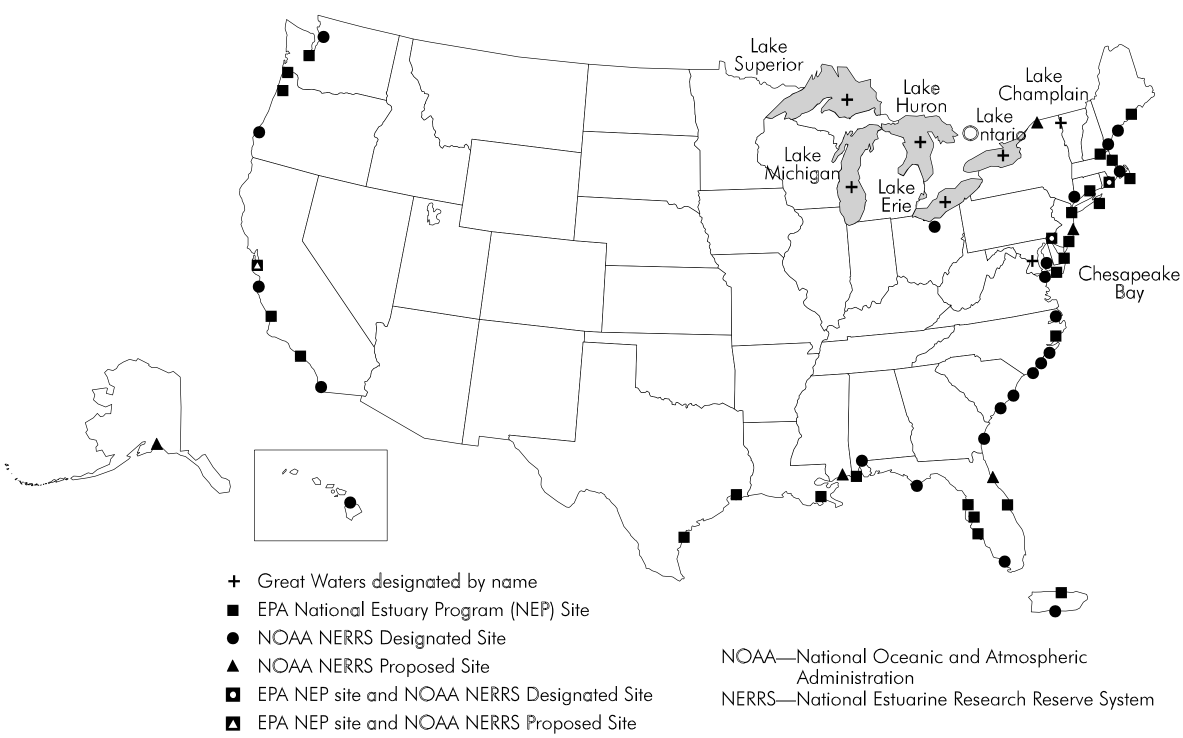
CHAPTER I
OVERVIEW OF THE GREAT WATERS PROGRAM

Section 112 of the Clean Air Act (CAA) provides the legislative basis for hazardous air pollutant (HAP) programs directed by the U.S. Environmental Protection Agency (EPA). In response to mounting evidence that air pollution contributes to water pollution, Congress included section 112(m), *Atmospheric Deposition to Great Lakes and Coastal Waters*, in the 1990 Amendments to the CAA to establish research, reporting, and potential regulatory requirements related to atmospheric deposition of HAPs to the "Great Waters." EPA coordinates activities to address the requirements of section 112(m) under the Great Waters program.

This report fulfills the requirements in section 112(m)(5), which directs EPA, in cooperation with National Oceanic and Atmospheric Administration (NOAA), to periodically submit a Report to Congress on atmospheric deposition to the Great Waters. The report is to describe "results of any monitoring, studies, and investigations conducted pursuant to" section 112(m). The First Report to Congress on atmospheric deposition to the Great Waters, referred to throughout this report as the "First Report to Congress," was published in May 1994 (U.S. EPA 1994a). This document is the Second Report to Congress and is intended to be an update of the information presented in the First Report to Congress.

The waterbodies collectively referred to as the "Great Waters" in this report are the Great Lakes, Lake Champlain, Chesapeake Bay, and specific coastal waters (i.e., defined in the statute as coastal waters designated through the National Estuary Program and the National Estuarine Research Reserve System). (See Figure I-1 for the locations of these waterbodies.)

FIGURE I-1
Locations of the Great Waters



I.A The Second Report to Congress

Goals of the Report

The main objective of this report is to update what is known about atmospheric deposition of pollutants to the Great Waters based on the scientific data available since publication of the First Report to Congress. The report focuses on research and activities in specific waterbodies to further understand and promote reductions of overall contaminant loadings to the Great Waters. In addition, this report includes a brief discussion of EPA's draft determination of the adequacy of section 112 to protect the Great Waters from deposition of HAP emissions from domestic stationary sources (see Chapter V).

EPA intends for this report to be an **update** of the First Report to Congress and has attempted to minimize restating information. In some instances, important findings or issues raised in the First Report to Congress are reiterated in this report to provide background information or to highlight an issue that continues to be significant to the Great Waters. For more detailed information on atmospheric deposition to the Great Waters, readers are encouraged to refer to the First Report to Congress (U.S. EPA 1994a), which summarized the scientific understanding of atmospheric deposition at that time and identified regulatory and research needs.

The scientific information presented in this report, together with the findings and recommendations identified in the First Report to Congress, should be used together to assess the progress since the First Report to Congress, and what data gaps still exist. Because of the short time period since the First Report to Congress, projects that were initiated after the release of the report or multi-year fieldwork projects, in most instances, are still in the data-gathering stage. Therefore, the results of these efforts cannot yet be analyzed. However, the objectives and status of these efforts are described in the report.

As in the First Report to Congress, the Second Report shows that data on effects, loadings, and sources are available to a certain extent; however, information to assess the linkage between these components remains inadequate, and therefore, unanswered questions, as well as uncertainties, persist for some of these issues. This report proposes a number of future directions (see Chapter V) to reduce uncertainty in several areas.

Report Preparation

The information in this report was collected from several sources. The references cited are generally from published peer-reviewed journals, government reports, and conference proceedings. The report uses sources that provide relevant information on Great Waters issues, but does not attempt to be comprehensive in the references cited. In general, literature published by late fall 1996 is included. Data on human health and ecological effects of pollutants of concern are based on a search for scientific literature published between completion of the background document on exposure and effects from the First Report to Congress (Swain et al. 1992a) through 1995. In a few instances throughout the report, more recent articles are included. In addition, in sections that are new to this report, older articles may be cited. Interested parties who know of other studies that may be pertinent to issues regarding atmospheric deposition to the Great Waters are encouraged to submit a copy of the article or a complete citation of the reference to EPA. Every effort will be made by EPA to review the article and to determine whether the information is relevant for the Third Report to Congress on atmospheric deposition to the Great Waters.

In addition to literature searches for current information on effects, current scientific information about atmospheric deposition to the Great Waters was compiled from two symposia held at the annual meeting of the Society of Environmental Toxicology and Chemistry (SETAC) in Denver, Colorado, from October 31 through November 4, 1994. Invited researchers presented findings from current research relevant to the Great Waters program. These findings have been assembled in a book entitled *Atmospheric Deposition of Contaminants to the Great Lakes and Coastal Waters* (Baker 1997).[1] In addition, EPA incorporated findings from recent investigations that have been funded by and/or conducted in connection with the Great Waters program, and has integrated findings from other significant EPA projects such the reassessment of dioxin and dioxin-like compounds (U.S. EPA 1994c, 1994d). Much of the waterbody-specific information presented in this report was provided by the EPA offices that coordinate investigation, restoration, and maintenance efforts in that waterbody (e.g., Great Lakes National Program Office, Chesapeake Bay Program Office).

The available waterbody-specific information on deposition of air pollutants focuses in large part on the Great Lakes and Chesapeake Bay. The Great Lakes have been a focus of the Great Waters Reports to Congress because, with their importance as the largest freshwater system in the United States and the observations reported for decades of toxic contamination in organisms living in the Great Lakes, there exists the best base of information on which to build. In addition, studies from the 1980s show atmospheric deposition to be a significant route of introducing pollutants to the Lakes. Knowledge gained of the conditions of the Great Lakes is useful in evaluating atmospheric deposition in other freshwaters as well. For example, researchers at Lake Champlain have developed scientific programs to determine the role of atmospheric deposition, particularly mercury, in water pollution. This report also focuses on Chesapeake Bay because accelerated eutrophication and its effects on the Bay have been recognized for over a decade. Accelerated eutrophication in the Bay is attributed, in part, to nitrogen loadings deposited from the atmosphere to the surrounding watershed, as well as directly into the Bay itself. Similar circumstances affect other U.S. estuaries, and information collected and applied in Chesapeake Bay will be useful for these waterbodies. For example, EPA has sponsored studies to refine the methodology used for estimating sources of nitrogen in Chesapeake Bay and to apply the methodology to estuaries in Galveston Bay, Texas, and Tampa Bay, Florida. A discussion of atmospheric deposition specific to each of the Great Waters — Great Lakes, Lake Champlain, Chesapeake Bay, and other coastal waters — is presented in Chapter IV.

The remaining sections of this chapter provide an overview of the findings from the First Report to Congress, as well as recent research activities in the Great Waters, followed by discussion of the air pollutants that are of particular concern to the Great Waters.

[1] Baker (1997) was still in press as of June 30, 1997.

I.B The First Report to Congress

The objective of the First Report to Congress was to describe what was known about atmospheric deposition of toxic chemicals to the Great Waters and present any appropriate regulatory recommendations based on the scientific information available at the time. The scientific content of the First Report to Congress was based mainly on three background documents prepared by committees of leading independent scientists (Baker et al. 1993; Keeler et al. 1993; Swain et al. 1992a). The information in these documents was used to answer three main scientific questions, develop scientific and policy conclusions, and recommend next steps. The three scientific questions were:

1. What human health and environmental effects are associated with the pollutants of concern in the Great Waters?

2. What is the relative importance of atmospheric deposition in causing contamination in the Great Waters?

3. What sources are significant contributors to atmospheric loadings to the Great Waters?

From the data compiled, three general responses to these questions were developed:

- Adverse effects (e.g., cancer, developmental effects) in humans and animals associated with exposure to the Great Waters pollutants of concern are fairly well understood. However, data are insufficient to establish a quantitative link between atmospheric deposition of these pollutants and their related effects.

- Atmospheric deposition can be a significant contributor of toxic chemicals to the Great Waters. The relative importance of atmospheric loading for a particular chemical in a given waterbody depends on characteristics of the waterbody, properties of the chemical, and the kind and amount of airborne, waterborne, and other sources.

- Many sources and source categories of pollutants of concern to the Great Waters have been identified. However, identification of particular sources responsible for the deposited pollutants in specific waterbodies is complicated since atmospheric loadings can originate from local, regional, and global sources.

Specific conclusions from the First Report to Congress, based on scientific data available at that time, included:

- Persistence and the tendency to bioaccumulate, critical characteristics of the Great Waters pollutants of concern, result in potentially greater human and ecological exposure to a pollutant in the environment.

- Exceedances of water quality criteria and standards have occurred for some of the pollutants in some waterbodies.

- ♦ Adverse effects on human health and wildlife have been observed due to exposure, especially through fish consumption, to persistent pollutants that bioaccumulate.

- ♦ In addition to cancer, noncancer effects (e.g., nervous system damage, immunological effects) caused by the pollutants can be a significant human health concern, and may affect some individuals exposed to levels above certain thresholds. Developing embryos and fetuses and breast-fed infants are given greater attention because they may be more susceptible than the general population to the adverse effects of these chemicals.

- ♦ Ecological effects on animal populations due to the pollutants of concern can be significant, such as immune function impairment, reproductive problems, and neurological changes that affect survival. Sometimes the effects on wildlife may be delayed and/or the symptoms subtle so that the effects are easily overlooked.

- ♦ Eutrophication resulting from excess nitrogen inputs is a major problem in some U.S. estuarine and coastal waters, and the relative contribution from atmospheric deposition of nitrogen to this problem can be significant. Ecological effects, ranging from nuisance algal blooms to oxygen depletion and fish kills, and adverse economic impacts to the waterbody region may result.

- ♦ Case studies have shown that atmospheric deposition can be a major contributor of mercury, nitrogen, polycyclic organic matter (POM), and polychlorinated biphenyls (PCBs) in waterbodies. The available information generally includes relative loadings estimates. Attention also should be given to the absolute quantity of the loadings because even small amounts of pollutants that bioaccumulate may produce a significant burden in fish and, ultimately, in humans and other fish-eating animals.

- ♦ Airborne emissions from both local and distant sources contribute to atmospheric deposition of pollutants to waterbodies. Deposition patterns can be influenced by characteristics of the pollutants and the source, and by weather and transport patterns.

- ♦ Continued research is needed, especially to help determine atmospheric contributions, to identify sources, to evaluate effects from low exposure levels, and to target HAPs that may pose the most significant risk to human health and aquatic resources.

Readers should refer to the First Report to Congress for discussion of the specific conclusions.

Based on the scientific conclusions in the First Report to Congress, EPA's principal policy conclusion was that reasonable actions are justified by the available scientific information, even though there are significant uncertainties associated with this information. While additional research is needed to reduce these uncertainties, reasonable actions to decrease atmospheric loadings need not wait for results of such information. To carry out its policy conclusion, EPA identified several recommendations for action, which were divided into three strategic themes:

1. EPA will continue ongoing efforts to implement section 112 and other sections of the CAA, as amended in 1990, and use the results of the First Report to Congress in the development of policy that will reduce emissions of the Great Waters pollutants of concern.

2. EPA recognizes the need for an integrated multimedia approach to the problem of atmospheric deposition of pollutants to waterbodies and, therefore, will utilize authorities beyond the CAA to reduce the human and environmental exposure to Great Waters pollutants of concern.

3. EPA will continue to support research activities and will develop and implement a strategy describing necessary research and policy assessments to address the mandates of section 112(m).

The specific action items based on these three strategic themes are described in detail in the First Report to Congress. The current status of each of the recommended action items is presented in Appendix A.

I.C Highlights of Progress Since the First Report to Congress

Much progress has been made since the First Report to Congress on research and other activities related to atmospheric deposition, especially activities that support section 112(m) mandates (see sidebar). The activities described in this report include those carried out by many national and regional EPA offices, as well as NOAA and a number of states (i.e., the programs and research were not all performed by the Great Waters program in EPA's Office of Air and Radiation). This report does not, however, attempt to be comprehensive in describing all the activities of these offices. A brief overview of some of the activities undertaken is presented below.

EPA Activities Addressing Section 112(m) Requirements

Section 112(m) directs EPA, in cooperation with NOAA, to identify and assess the extent of atmospheric deposition of toxic pollutants to the Great Waters. As part of the assessment, EPA supports the following activities:

- Monitoring of atmospheric deposition, including the establishment of monitoring networks in the Great Lakes, Chesapeake Bay, Lake Champlain, and coastal waters;
- Investigation of sources and deposition rates of air pollutants;
- Research for developing and improving monitoring methods and for determining the relative contribution of atmospheric pollutants to total pollutants in the Great Waters;
- Evaluation of adverse human health and environmental effects;
- Identification of exceedances of water quality and drinking water standards;
- Sampling of fish and wildlife for pollutants of concern;
- Characterization of sources of pollutants of concern; and
- Determinations of whether section 112 authority is "adequate to prevent serious adverse effects to public health and serious or widespread environmental effects" associated with atmospheric deposition of HAPs to the Great Waters, and of whether further emissions standards or control measures to prevent such effects are necessary and appropriate. Based on these determinations, EPA is directed to take additional measures, as necessary and appropriate, to prevent such adverse effects to human health and the environment.

♦ EPA has worked with the Great Lakes States to continue development of regional emission inventories for the Great Lakes and a data storage and retrieval system. Data collection was recently completed, and the data base will be updated annually. Work will continue to characterize mobile source emissions and to improve the accuracy of the emissions inventory. Determining, categorizing, and estimating the magnitude of pollutant sources will be a significant step toward reduction of atmospheric loading of pollutants to the Great Lakes.

- Quantitative data continue to be gathered on atmospheric deposition of pollutants including PCBs, DDT, dieldrin, and lindane in each of the Great Lakes through the Integrated Atmospheric Deposition Network (IADN) (a joint U.S./Canadian monitoring network). Recent data have been incorporated into deposition estimates for 1994, thereby allowing comparison of data to 1992 results.

- Atmospheric mercury concentration and deposition have been monitored continuously in the Lake Champlain region in the last few years, which will be important for determining atmospheric deposition trends in the lake basin.

- A large-scale airshed model for Chesapeake Bay has been developed to determine the general geographical location and type of sources of nitrogen emissions, and the relative contributions of different sources and patterns of nitrogen deposition to the Bay watershed and directly to tidal surface waters. Models of the Chesapeake Bay airshed, watershed, and tidal waters were extensively revised to link daily atmospheric deposition loading data to models of water quality impacts in the tidal Bay and resultant influences on Bay underwater grasses, bottom benthic communities, and overlying fish habitat.

- The Chesapeake Bay Atmospheric Deposition Study (CBADS) network was established and the resultant data have been used to quantify atmospheric loadings and depositional fluxes of toxic contaminants to the Bay, as part of the development of a larger basinwide chemical contaminant loading and release inventory.

- Research stations have been established to measure atmospheric deposition of nitrogen and other selected pollutants in Tampa Bay, Galveston Bay, and Pamlico Sound and adjacent estuaries (e.g, Neuse River Estuary, Newport River Estuary); collected data will be used to determine annual atmospheric loadings of these pollutants and the relative contribution of remote and local sources to atmospheric deposition in the waterbodies.

Chapter IV of this Report to Congress provides more details on the activities highlighted above.

I.D Pollutants of Concern

Great Waters Pollutants of Concern and Reasons for Inclusion

As did the First Report to Congress, this report focuses on selected pollutants of concern (see sidebar on next page). These pollutants are potentially of concern for atmospheric deposition to the Great Waters.[2] The general types of sources and uses (and use restrictions) of these pollutants are briefly summarized in Table I-1.

The list of 15 Great Waters pollutants of concern has not been expanded since the First Report to Congress. Three pesticides, atrazine, hexachlorobutadiene, and methoxychlor, mentioned in the First Report, continue to be considered by EPA as potential future additions to the Great Waters list of pollutants of concern. Atrazine warrants continued attention as a potential pollutant of concern because of its widespread occurrence (e.g., commonly used in the Great Lakes basin), its at-least moderate persistence, and its potential to cause a variety of effects on biota. For these reasons, atrazine is also one of the chemicals of focus for the Lake Michigan Mass Balance Study (discussed in Chapter IV). The other two pesticides under consideration for future addition, hexachlorobutadiene and methoxychlor, are both on the CAA HAPs list and have the potential to bioaccumulate in the food web. Additional information suggests that atrazine and methoxychlor are potential endocrine disruptors, a group of chemicals that mimic or otherwise interfere with hormones in the body, resulting in various adverse biological effects.

Great Waters Pollutants of Concern

Cadmium and cadmium compounds
Chlordane
DDT/DDE
Dieldrin
Hexachlorobenzene (HCB)
α-Hexachlorocyclohexane (α-HCH)
Lindane (γ-hexachlorocyclohexane; γ-HCH)
Lead and lead compounds
Mercury and mercury compounds
Polychlorinated biphenyls (PCBs)
Polycyclic organic matter (POM)
Tetrachlorodibenzo-p-dioxin, 2,3,7,8- (TCDD; dioxins)
Tetrachlorodibenzofuran, 2,3,7,8- (TCDF; furans)
Toxaphene
Nitrogen compounds

<u>Under Evaluation for Addition to Great Waters List</u>
Atrazine
Hexachlorobutadiene
Methoxychlor

The 15 pollutants of concern for the Great Waters were selected based on available data on their effects and deposition. Reasons for selecting these pollutants include:

- All the pollutants, except for nitrogen compounds, persist in the environment and/or have a high potential to accumulate in living organisms. All the pollutants can cause adverse effects in humans and the environment.

- All 15 pollutants are known air pollutants and are known to be present in atmospheric deposition (e.g., rainfall, dry deposition).

- Data indicate that these pollutants are present in the waters and biota of the Great Waters and that one route of pollutants to these waterbodies is atmospheric deposition.

[2] The pollutants of concern are not considered to be inclusive of *all* chemicals that may, now or in the future, be an important component of atmospheric deposition to the Great Waters. While nitrogen is not listed as a HAP under section 112(b) of the CAA, this report examines the contribution of excess levels of nitrogen to eutrophication. Acidification or "acid rain" is not discussed because it is addressed under a separate CAA program.

TABLE I-1
Pollutants of Concern in the Great Waters[a]

Pollutant	Examples of Uses[b]
Cadmium and compounds	Naturally occurring element used in metals production processes, batteries, and solder. Often released during combustion of fossil fuels and waste oil, and during mining and smelting operations.
Chlordane	Insecticide used widely in the 1970s and 1980s. All U.S. uses except termite control cancelled in 1978; use for termite control voluntarily suspended in 1988. Use of existing stocks permitted.
DDT/DDE	Insecticide used widely from introduction in 1946 until significantly restricted in U.S. in 1972. Still used in other countries. Used in U.S. for agriculture and public health purposes only with special permits.
Dieldrin	Insecticide used widely after introduction in late 1940s. Used in U.S. for termite control from 1972 until registration voluntarily suspended in 1987.
Hexachlorobenzene	Fungicide used as seed protectant until 1985. By-product of chlorinated compound and pesticide manufacturing. Also a by-product of combustion of chlorine-containing materials. Present as a contaminant in some pesticides.
α-Hexachlorocyclohexane (α-HCH)	Component of technical-HCH, an insecticide for which use is restricted in U.S., but which is used widely in other countries.
Lindane (γ-Hexachlorocyclohexane; γ-HCH)	An insecticide used on food crops and forests, and to control lice and scabies in livestock and humans. Currently used primarily in China, India, and Mexico. U.S. production stopped in 1977. Use was restricted in 1983; however, many uses are still registered, but are expected to be voluntarily cancelled in the future.
Lead and compounds	Naturally occurring element commonly used in gasoline and paint additives, storage batteries, solder, and ammunition. Released from many combustion and manufacturing processes and from motor vehicles. Use in paint additives restricted in U.S. in 1971. U.S. restrictions on use in gasoline additives began in 1973 and have continued through the present, with a major use reduction in the mid-1980s.
Mercury and compounds	Naturally occurring element often used in thermometers, electrical equipment (such as batteries and switching equipment), and industrial control instruments. Released from many combustion, manufacturing, and natural processes. Banned as a paint additive in U.S. in both interior (1990) and exterior (1991) paint.
Polychlorinated biphenyls (PCBs)	Industrial chemicals used widely in the U.S. from 1929 until 1978 for many purposes, such as coolants and lubricants and in electrical equipment (e.g., transformers and capacitors). In the U.S., manufacture stopped in 1977 and uses were significantly restricted in 1979. Still used for some purposes because of stability and heat resistance, and still present in certain electrical equipment used throughout the United States.
Polycyclic organic matter (POM)[c]	Naturally occurring substances that are by-products of the incomplete combustion of fossil fuels and plant and animal biomass (e.g., forest fires). Also, by-products from steel and coke production and waste incineration.
TCDD (dioxins)	By-product of combustion of organic material containing chlorine, chlorine bleaching in pulp and paper manufacturing, and diesel-fueled vehicles. Also a contaminant in some pesticides.
TCDF (furans)	By-product of combustion of organic material containing chlorine, chlorine bleaching in pulp and paper manufacturing, and diesel-fueled vehicles. Also a contaminant in some pesticides.
Toxaphene	Insecticide used widely on cotton in the southern U.S. until the late 1970s. Most U.S. uses banned in 1982; remaining uses cancelled in 1987.
Nitrogen compounds	By-products of power generation, industrial, and motor vehicle fossil fuel combustion processes (NO_x and NH_3). Also, compounds used in fertilizers and released from agricultural animal manures (NH_3).

[a] Source: See the First Report to Congress for references for this table (U.S. EPA 1994a).
[b] Applicable restrictions (including bans) on use or manufacture in United States also are described.
[c] POM is a large class of chemicals consisting of organic compounds having multiple benzene rings and a boiling point greater than 100° C. Polycyclic aromatic hydrocarbons (PAHs) are a chemical class that is a subset of POM.

- These pollutants overlap substantially with the toxic air pollutants that ranked highest in an EPA-sponsored study (ICF 1991) to identify priority chemicals having characteristics that lead to potential adverse effects in the Great Waters.

- With the exception of dieldrin and nitrogen compounds, all of these pollutants are listed as HAPs under section 112(b) of the CAA.[3]

- With the exception of 2,3,7,8-TCDF and nitrogen compounds, these pollutants are included on the list of pollutants that were the initial focus of the EPA/state Great Lakes Water Quality Initiative.[4] They are considered to be potentially significant as air pollutants deposited to the Great Lakes.

- Ten of the 15 pollutants are designated as bioaccumulative chemicals of concern under EPA's Great Lakes Water Quality Guidance.[5]

- Six of the 15 pollutants (cadmium, chlordane, lead, mercury, PCBs, and several PAHs (which are part of the POM class of compounds)) are on the Chesapeake Bay Toxics of Concern List,[6] and two more pollutants, dieldrin and toxaphene, are listed as potential future additions to this list.

- Nitrogen compounds play an important role in excessive nutrient enrichment in many estuaries and coastal waters, and numerous studies indicate that atmospheric loadings of nitrogen to the Chesapeake Bay and other coastal waters are a significant portion of total nitrogen loadings. In most freshwaters, nitrogen compounds play a less immediate role in promoting excessive enrichment. For example, airborne nitrogen compounds are not of concern currently in the Great Lakes.

- The pollutant list overlaps substantially with several sets of Great Lakes chemicals of concern selected by other scientific and regulatory groups, including the Great Lakes Water Quality Board (GLWQB) of the International Joint Commission (IJC), a cooperative committee comprised of U.S. and Canadian representatives.

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[3] Several pollutants of concern are listed by a different name in section 112(b). The pollutants of concern are listed in section 112(b) as: cadmium compounds, chlordane, DDE, hexachlorobenzene, lindane (all isomers, which includes α-HCH), lead compounds, mercury compounds, PCBs, POM, 2,3,7,8-TCDD, dibenzofurans, and toxaphene. In addition, hexachlorobutadiene and methoxychlor are listed in section 112(b).

[4] Established in 1989 to a provide consistent level of environmental protection for the Great Lakes ecosystem, this Initiative supported principles and goals of the 1986 Great Lakes Toxic Substances Control Agreement (Governors' Agreement).

[5] The Final Water Quality Guidance for the Great Lakes System was released in 1995 (60 *Federal Register* 15366) and resulted in the deletion of six chemicals (including aldrin, endrin, methoxychlor) from the proposed 1993 list of bioaccumulative chemicals of concern. The final guidance also eliminated the list of 10 pollutants considered potential bioaccumulative chemicals of concern. Although furans (2,3,7,8-TCDF) are not specified in the 1995 guidance, criteria for furans may eventually be set.

[6] Atrazine is also found on the Chesapeake Bay Toxics of Concern List.

Use of Pollutant Groups

In some sections of this report, discussion of the Great Waters pollutants of concern is organized by pollutant group. The five pollutant groups used in this report are described below. Many of the pollutants may fit into more than one group, but have been placed in the most appropriate category.

EPA has organized the pollutants of concern in these five groups for several reasons. First, the pollutants in each group generally originate from similar sources or are released through similar mechanisms. Thus, action proposed to reduce emissions of individual pollutants may be applied more broadly to the entire group. Second, pollutants in each group may have similar chemical characteristics, allowing for generalizations related to deposition and cycling within the environment. Third, separating the pollutants into various groups allows for pollutants with unique regulatory concerns, such as mercury and nitrogen, to be highlighted and emphasized in the Report to Congress. Finally, grouping the pollutants helps decision-makers develop conclusions about pollutants with similar chemical/physical behavior or sources, where there are limited data.

- **Mercury and mercury compounds**. Mercury is released as an air pollutant from a variety of natural and anthropogenic area and point sources (including combustion and manufacturing sources). Although mercury is a metal, it is treated in this report as a separate pollutant group because it behaves differently in the environment than other metals and produces different types of effects, as well as because of the comprehensive data that are available for it. Mercury can be found in elemental, inorganic, or organic forms in the environment. In aquatic species, mercury exists primarily as organic mercury (e.g., methylmercury), which can bioaccumulate in tissues and biomagnify in the food web. In addition, special emphasis is given to mercury emissions in the CAA. Several subsections of section 112 require studies to be conducted on mercury as a toxic air pollutant; a review draft of an EPA report related to atmospheric emissions of mercury was submitted to the Science Advisory Board (SAB) in 1996. When submitted to Congress, the final Mercury Study Report will fulfill the mandate under CAA section 112(n)(1)(B) that the study consider:

    - The rate and mass of mercury emissions;
    - The health and environmental effects of such emissions;
    - Technologies that are available to control such emissions; and
    - The cost of these control technologies.

- **Other metals**. Cadmium compounds and lead compounds comprise this group. These metal compounds are released from various combustion and production processes. Note, however, that a significant source of lead was reduced following the phaseout of lead in gasoline additives that began in the early 1970s.

- **Combustion emissions**. The pollutants in this group include PCBs, POM, 2,3,7,8-TCDD, and 2,3,7,8-TCDF. (See the sidebar on the next page for a discussion of TCDD and TCDF.) These pollutants generally are released during incomplete combustion of fossil fuels and/or combustion during manufacturing or incineration processes. PCBs, though historically used in electrical equipment and hydraulic fluids, are included in this group because they may be released to the atmosphere in combustion gases when PCB-containing materials are burned.

- **Pesticides**. This group includes chlordane, DDT/DDE, dieldrin, hexachlorobenzene, α-HCH, lindane, and toxaphene. Although the use of these pesticides is significantly limited in the United States, they continue to be of concern in the Great Waters because of their persistence in the environment and the long-range transport from other countries in which the pesticides are still used. Atrazine, hexachlorobutadiene, and methoxychlor are potential future additions to this group.

- **Nitrogen compounds**. This group includes nitrogen oxides, reduced nitrogen compounds (such as ammonia and ammonium), and organic nitrogen. These pollutants are released through both natural and anthropogenic pathways. Although nitrogen oxides are fossil fuel combustion by-products, nitrogen compounds are treated as a separate pollutant group because: (1) other measures are being taken to control nitrogen through programs related to ground-level ozone and acid precipitation; (2) nitrogen, unlike the other selected pollutants of concern, is an essential nutrient and is not listed as a HAP under CAA section 112(b); and (3) when present in excessive amounts, nitrogen (in oxides and other compounds that are plant nutrients) is the nutrient driving the accelerated eutrophication of most estuarine and coastal waters, resulting in significant adverse ecosystem effects. Unlike the other pollutants, nitrogen is a required nutrient that supports the ecosystem and becomes a pollutant when it reaches levels that result in overfertilization with deterioration of water quality.

Dioxins and Furans

Section 112(b) of the CAA includes in its list of HAPs "2,3,7,8-tetrachlorodibenzo-*p*-dioxin" and "dibenzofuran." These two substances are part of a much larger class of compounds, as discussed below.

Dibenzo-*p*-dioxin and dibenzofuran molecules both carry single hydrogen atoms bonded to carbon atoms at the outside corners. When chlorine atoms are substituted for any (or all) of these hydrogens, the compounds become chlorinated dibenzo-*p*-dioxins and chlorinated dibenzofurans (CDDs and CDFs). The presence of chlorine may increase the toxicity of the compound by many orders of magnitude, depending on their number and location. There are 75 possible CDD compounds and 135 possible CDF compounds. Each of these individual CDD and CDF forms is called a "congener." It is these CDD and CDF congeners that are commonly referred to as "dioxins" and "furans." The terms "dioxins" or "TCDD" and "furans" or "TCDF" are used in this report to refer to all CDD and CDF congeners, respectively.

Relationship of Pollutants of Concern to Section 112 and Other CAA Requirements

Table I-2 presents the section 112 requirements that may regulate emissions of each pollutant of concern. As shown, emissions of mercury are covered most comprehensively by section 112 requirements, followed by emissions of lead compounds, POM, TCDD, and TCDF. (Emissions of lead compounds also are regulated under the CAA Title I criteria air pollutant program.) Emissions of hexachlorobenzene and PCBs may be regulated under the maximum achievable control technology (MACT) standards required by sections 112(d), (g), and (j), and under 112(c)(6). Emissions of cadmium compounds are covered under the MACT standards and, for electric utility steam generating units, under 112(n)(1)(A). For several pesticides, the development of MACT standards is the main section 112 requirement that may control emissions of these pollutants, to the extent that major sources of air emissions still exist in the United States. Section 112(f), which is not included in the table, is intended to address the public health risks and adverse environmental effects from HAP

TABLE I-2
Great Waters Pollutants of Concern and CAA Section 112

Pollutant	Applicable CAA Section 112 Activities[a]					
	112(b)	112(c)(6)	112(d),(g),(h),(j)	112(n)(1)(A)	112(n)(1)(B)	112(n)(1)(C)
Cadmium and compounds	●		●	●		
Chlordane	●		●			
DDT/DDE	●		●			
Dieldrin						
Hexachlorobenzene	●	●	●			
α-HCH	●		●			
Lead and compounds[b]	●	●	●	●		
Lindane	●		●			
Mercury and compounds	●	●	●	●	●	●
PCBs	●	●	●			
Polycyclic organic matter (POM)	●	●	●	●		
TCDD (dioxins)	●	●	●	●		
TCDF (furans)	●	●	●	●		
Toxaphene	●		●			
Nitrogen compounds[c]						

[a] 112(b) = HAP list; the pollutants of concern are regulated under section 112 only by their name as listed in section 112(b) (cadmium compounds, chlordane, DDE, hexachlorobenzene, lindane (all isomers, which includes α-HCH), lead compounds, mercury compounds, PCBs, POM, 2,3,7,8-TCDD, dibenzofurans, and toxaphene).
112(c)(6) = Sources accounting for 90 percent of these emissions to be subject to regulation
112(d),(g),(h),(j) = Emissions of HAPs from major sources subject to regulation by MACT standards
112(n)(1)(A) = Emissions of these six HAPs from electric utility steam generating units to be evaluated for regulation
112(n)(1)(B) = Emissions of mercury from electric utilities, municipal waste combustors, and other sources to be studied
112(n)(1)(C) = Report required on "threshold" level for human health effects from mercury
Does not include section 112(f), which is intended to address the public health risks and adverse environmental effects from HAP emissions remaining after implementation of 112(d) standards.
[b] Lead compounds also regulated under the criteria air pollutant program.
[c] Nitrogen oxides (NO_x) regulated under several other CAA programs, such as those that control criteria air pollutants, mobile source emissions, and acid rain.

emissions remaining after implementation of section 112(d) MACT standards; these standards could apply to any HAP for which it is determined that "residual risk" remains. Emissions of dieldrin and nitrogen compounds are not currently addressed by the section 112 requirements listed in Table I-2. Emissions of certain nitrogen compounds, however, are regulated under several CAA programs other than section 112, including the Title I criteria air pollutant program, the Title II mobile sources program, and the Title IV acid deposition program.

Other sections of the CAA may also regulate emissions of the pollutants of concern. For example, under section 129 of the CAA, which applies to municipal waste combustors, EPA is to develop numerical emission limitations for several pollutants, including the following Great Waters pollutants of concern: cadmium, dioxins, furans, lead, mercury, and nitrogen compounds (nitrogen oxides).

CHAPTER II
EXPOSURE AND EFFECTS

Section 112(m) of the CAA requires EPA to assess the environmental and human health effects attributable to atmospheric deposition to the Great Waters and to assess whether atmospheric pollutant loadings to the Great Waters cause or contribute to exceedances of drinking water or water quality standards.

Adverse effects on environmental and human health and exceedances of drinking water or water quality standards that result from the pollutants of concern are pieces of a larger puzzle of what happens to the pollutants of concern after they are deposited to the Great Waters. After being deposited to water, the pollutants can bind to particles, concentrate at the water surface, dissolve in the water, and/or (if sufficiently volatile) escape as gases back into the air. Ecosystems and humans may be exposed to these pollutants through various exposure routes (e.g., food consumption). Exceedances of water quality criteria or standards are one means of assessing the levels of the pollutants in water and biota to which ecosystems and humans may be exposed. Following exposure to the pollutants of concern, ecosystems and humans may experience adverse health effects.

At this time, it is not possible to distinguish between effects caused by airborne pollutants and the same pollutants delivered by waterborne or other routes. In the absence of data to the contrary, EPA takes the position that the contribution of airborne pollutants to adverse effects corresponds to the relative air contribution by various routes to pollutant loads, including releases of historic loadings from sediments.

This chapter is divided into five sections that discuss:

- Exposure routes and extent of contamination in the Great Waters (Section II.A);
- Level of contamination in biota of the Great Waters (Section II.B);
- Potential ecological effects that may result from exposure to the pollutants of concern (Section II.C);
- Potential human health effects that may result from exposure to the pollutants of concern (Section II.D); and
- Other potential effects, such as recreational fishing losses, attributable to the pollutants of concern (Section II.E).

Sections II.A, II.C, and II.D update information that was presented in the First Report to Congress (U.S. EPA 1994a). These sections begin with a brief summary of the information presented in the First Report to Congress to provide a foundation for the subsequent discussion of the recent information available for this report.

II.A Exposure Routes and Extent of Contamination

This section presents information on the exposure routes of concern for humans and ecosystems and the extent of contamination in the Great Waters. The measures used in this report to assess pollutant exposure and the extent of contamination are exceedances of water quality criteria and the issuance of fishing restrictions and fish consumption advisories. Levels of contamination in biota are discussed separately in Section II.B.

As noted earlier, the relationship between exposure and resulting adverse effects of toxic pollutants and atmospheric deposition is not well understood. As described in the First Report, some correlations and linkages have been established between specific pollutants of concern and exposure and effects in the Great Waters. Many pollutants that are a concern due to atmospheric deposition also have a long history of direct surface water discharges to the Great Waters. In addition, current pollutant levels in waterbodies may include the contribution of pollutants that enter through groundwater, that are recycled from sediments, or that are resuspended, following earlier deposition, and redeposited at other locations. There currently is no evidence available to suggest that the pollutants deposited from the air will have effects on biota any different from the effects of these same pollutants carried in water or found in sediment.

Conclusions from the First Report to Congress

As mentioned above, information from the First Report to Congress is presented here to provide a foundation for the subsequent discussion of the recent information available for the Second Report. The research findings and studies presented in the First Report, as well as the background document on exposure and effects (Swain et al. 1992a), led to the following conclusions concerning exposure routes and extent of contamination in the Great Waters:

- For water pollutants that are derived from atmospheric deposition, the major routes of exposure are fairly well understood. The main exposure routes of concern for animals are intake of food, intake of drinking water, and direct contact with water. Exposure routes for plants include water uptake and direct contact. For humans and fish-eating birds and mammals, intake of food (e.g., contaminated fish) is the main exposure route of concern for pollutants that are persistent in the environment and that tend to bioaccumulate.

- The pollutants of concern generally are persistent in the environment and tend to accumulate in fat or muscle tissue and, as a result of food web interactions, reach the highest concentrations in animals at the top of the food web, including humans. These characteristics allow the pollutants to remain in the environment and animal tissue for long periods of time, increasing the opportunity for exposure and resulting in greater exposures to animals at the top of the food web.

- The tendency of the pollutants of concern to bioconcentrate and biomagnify was supported by numerous studies summarized in the First Report. (See sidebar on next page for definition of terms. Note that these terms may be used with somewhat different emphasis by different authors. This summary is based on common usages from many articles reviewed for this report.) Evidence presented by these studies included: (1) documented cases of elevated levels of persistent toxic pollutants in various fish species compared to levels in water and, in many cases, levels in sediment; (2) data showing that predators (e.g., the herring gull, bald eagle, and turtle) in the Great Lakes region have had some of the highest reported concentrations of persistent toxic chemicals in their tissues; and (3) data indicating that people who regularly consumed fish from Lake Michigan in the 1970s had significantly higher concentrations of PCBs and pesticides, such as DDT, in their tissues compared with those who did not consume fish.

Distinguishing Common Terms Describing Bioaccumulation

Bioaccumulation is the uptake and retention of a chemical by a living organism as a result of intake of food, intake of drinking water, direct contact, or inhalation.

Bioconcentration is the phenomenon by which chemicals become more concentrated in an organism than in its surrounding environment.

Biomagnification is the phenomenon by which chemicals become more concentrated in animals at higher levels in the food web.

- Based on the most current information available for the First Report to Congress, portions of all of the Great Lakes and many associated waterbodies, Chesapeake Bay, and Lake Champlain had some kind of advisory on fish consumption at that time.

- Understanding of the contribution of atmospheric deposition to overall exposure was limited for the First Report to Congress because: (1) overall exposure to toxic water pollutants had not been adequately quantified; (2) sufficient and accurate information on all pollutant inputs and outputs was not available at that time; and (3) the difficulty in distinguishing the origin of a pollutant (e.g., originated from the air) after it is in the water made it difficult to link exposure, and resultant effects, to particular pathways (e.g., atmospheric deposition).

- Although the exposure routes of concern have been identified, the concentrations of pollutants in water to which humans, animals, and plants are exposed (i.e., the extent of contamination) were not easily determined given available data at that time.

- Few violations of existing drinking water standards (i.e., maximum contaminant levels or MCLs) for the pollutants of concern were found in Great Lakes drinking water systems; for the pollutants that exceeded their MCLs, much of the problem was thought to be caused by the distribution system rather than the water source.

- When maximum open water concentrations from Great Lakes sampling data taken between 1980-1986 were compared to water quality criteria, six pollutants of concern (cadmium, dieldrin, DDT/DDE, hexachlorobenzene, mercury, and PCBs) potentially exceeded at least one criterion in at least one of the Great Lakes (see Appendix B of the First Report to Congress). Maximum concentrations of most of the remaining Great Waters pollutants in most of the lakes approached levels of concern. An updated comparison of sampling data to water quality criteria is presented later in this section.

- In Lake Champlain, limited sampling data indicated that lead was the only pollutant of concern that exceeded applicable water quality criteria. In Chesapeake Bay, a limited number of measured concentrations of cadmium and lead in the tidal tributaries to the Chesapeake Bay exceeded EPA water quality criteria and state water quality standards prior to 1993.

The remainder of this section presents updated information on exposure routes and extent of contamination.

Current Understanding of Exposure Routes and Extent of Contamination

As indicated above and in the First Report to Congress, the exposure routes of concern for humans and ecosystems are fairly well understood. Exposures can occur through intake of drinking water, direct contact with water, and, especially important for humans and fish-eating birds and mammals, intake of food. This section presents updated information on measures for assessing the extent of contamination in the Great Waters.

COMPARISON TO WATER QUALITY CRITERIA

One means of assessing the extent of contamination in the Great Waters caused by the pollutants of concern is to compare available water sampling data to drinking water standards and other water quality criteria. Such comparisons are consistent with the requirement in section 112(m) of the 1990 CAA Amendments for EPA to assess the contribution of atmospheric deposition to exceedances of drinking water standards and other water quality standards and criteria.

For national drinking water standards, few exceedances are known for the Great Waters pollutants of concern based on current information in the Great Lakes. Since the First Report to Congress, there continue to be few violations of existing maximum contaminant levels (MCLs) in Great Lakes drinking water systems. Interpretation of this information is limited because the exceedance of the MCL for a pollutant may be a result of a problem in the distribution system, rather than the water source.

For other water quality criteria and standards, three sets of relevant water quality criteria are compared with available Great Waters sampling data: EPA's national ambient water quality criteria (AWQC); the U.S.-Canadian Great Lakes water quality objectives (GLWQOs); and Great Lakes water quality criteria (GLWQC) developed by EPA and Great Lakes states. The first two sets of criteria are the same as those used in the First Report, while the third set, GLWQC, was released in 1995. Proposed GLWQC (pGLWQC) were used in the First Report, but these criteria have since been finalized (U.S. EPA 1995a); see Chapter IV for more discussion on the development of GLWQC. The three sets of criteria are briefly defined in Table II-1.

Water sampling data are compared with water quality criteria, rather than comparing sediment contamination data or biological contamination data to appropriate standards, for two main reasons: (1) the specific requirement in section 112(m) to report exceedances of water quality standards and benchmarks, and (2) the limited availability of federal or other widely accepted numerical benchmarks for sediments or living organisms for the selected pollutants of concern. Because many of the pollutants of concern bioconcentrate and biomagnify, water concentrations may understate the full potential for fish and wildlife to contain high concentration levels; only the GLWQC account for the potential for biomagnification. Therefore, the absence of water quality criteria exceedances for pollutants that have a strong tendency to bind to sediments and to bioaccumulate does not necessarily indicate the absence of contamination levels of potential human health or ecological concern. Contamination levels in biota and sediments in the Great Waters are discussed in Section II.B.

Table II-2 compares recent estimates of total water column concentrations in the Great Lakes for seven pollutants of concern for which sampling data exist (i.e., DDT/DDE, dieldrin, hexachlorobenzene (HCB), α-HCH, lindane, PCBs, and POM). The data are taken from studies conducted between 1986 and 1991 by EPA, researchers funded by EPA, and Environment Canada. Sampling data for the other pollutants of concern and for other Great Waters were not readily available.

TABLE II-1
Summary of Water Quality Criteria Used for Comparison in This Report

Criteria Set[a]	Summary
Ambient water quality criteria (AWQC)	Designed to protect humans, and freshwater and saltwater animals and plants from harmful effects resulting from chronic and acute exposures. Reflect current knowledge on health and welfare effects, dispersal of pollutants across media, and effects on animal and plant reproduction and communities. Derived entirely with risk-based data (not cost or technology considerations). National criteria provided by EPA as guidelines to states for developing regulations.
Great Lakes water quality objectives (GLWQOs)	Developed through joint U.S.-Canadian agreement. Set for certain chemicals to protect the most sensitive user of the water among humans, aquatic life, and wildlife. For chemicals with no specific GLWQO, concentrations in water (does not specify whether ambient water) and in aquatic organisms should be lower than detection levels.
Great Lakes water quality criteria (GLWQC)	Developed by EPA and Great Lakes States. Specific to the Great Lakes system. Form basis for new state water quality standards for ambient waters of the Great Lakes system. Provided as guidelines to protect aquatic life (for both acute and chronic exposure), wildlife (for exposure through food webs), and humans (for chronic exposure through consumption of both fish and drinking water and through water-related recreation). Includes consideration of biomagnification.

[a] Sources: U.S. EPA 1986, IJC 1978, and U.S. EPA 1995a, respectively.

As shown in Table II-2, total water column concentrations of dieldrin and PCBs exceed their GLWQC at some locations in all of the Great Lakes, where data are available. Note that DDT/DDE may also exceed its GLWQC (sampling data of $<0.00006\ \mu g/L$ versus GLWQC of $0.000011\ \mu g/L$), and that the criterion for POM is for polycyclic aromatic hydrocarbons (PAHs), a subset of POM, while sampling data were available only for one POM compound (i.e., benzo(a)pyrene, or B(a)P). In addition, the concentrations of PCBs at some locations in Lakes Erie, Huron, and Ontario are above the AWQC for human health. For the pollutants with sampling data reported in both the First Report to Congress and this report (i.e., dieldrin, DDT/DDE, HCB, and PCBs), the total water column concentrations presented in Table II-2 are generally lower than the concentrations reported in the First Report to Congress and the levels for DDT/DDE and HCB no longer exceed any of the water quality criteria.

TABLE II-2
Comparison of Water Quality Criteria to Pollutant Concentrations in the Great Lakes (μg/L)

Pollutant	National AWQC: Fresh Water Aquatic Life[a]	National AWQC: Human Health[b]	Great Lakes Water Quality Agreement Objective[c]	Great Lakes Water Quality Criterion[d]	Total Water Column Concentration[e]				
					Lake Superior	Lake Michigan	Lake Huron	Lake Erie	Lake Ontario
DDT/DDE[f]	0.001	0.00024	0.003	0.000011	<0.00006	NA	<0.00006	<0.00006	<0.00006
Dieldrin	0.0019	0.00071	0.001[g]	0.0000065	0.00026	NA	0.00032-0.00035	0.00038	0.00028-0.00032
HCB	—	0.0072	—	0.00045	<0.00004	NA	0.000072	0.000047	0.000036
α-HCH	—	0.092	—	—	0.0011	0.0016	0.0015	0.0011	0.0008-0.0009
Lindane	0.08	0.186	0.01	0.47	0.0004	0.00034	0.00038	0.00049	0.00036
Total PCBs	0.014	0.00079	—	0.0000039	0.00018	0.00020-0.00036	0.0007-0.0009	0.00122	0.0012
POM[h]	—	0.028	—	—	<0.00046	NA	<0.00046	<0.00046	<0.00046

NA=No data available.
Highlighted boxes indicate exceedances of GLWQC; shaded boxes indicate exceedances of AWQC for human health.

[a] Values are for freshwater chronic criteria (U.S. EPA 1986).
[b] Values are for human chronic exposure through both fish consumption and drinking water (U.S. EPA 1986).
[c] Values are for protection of the most sensitive user of the water among humans, aquatic life and wildlife (IJC 1978).
[d] Values are the most stringent (i.e., lowest) among those for protection of human health, aquatic life, or wildlife (U.S. EPA 1995a).
[e] Concentrations are taken from De Vault et al. (1995) and L'Italien (1993). Concentrations of dieldrin and PCBs that are reported as ranges represent two different concentrations reported in two different studies. For α-HCH, the range of concentrations in Lake Ontario represents the range reported in a single study.
[f] Sampling data are for p,p'-DDE.
[g] Value for aldrin and dieldrin combined.
[h] AWQC for human health is for polycyclic aromatic hydrocarbons (PAHs), a subset of POM; sampling data are for benzo(a)pyrene, a PAH.

Meaningful trend data on pollutant concentrations in the Great Lakes are limited mainly because the technology required to measure pollutants at the trace concentrations found in the water column of the Great Lakes has become widely available only in the last few years (De Vault et al. 1995). One recent study, however, has provided insight into water column trends for PCBs in Lake Superior (Jeremiason et al. 1994). As shown in Table II-3, total PCB concentrations in the Lake Superior water column show an overall decline from 1978 to 1992, though there is some variation from year to year. In addition, based on the concentrations of the same 25 PCB congeners measured in surface water samples collected between 1980 and 1992 by the same laboratory (also shown in Table II-3), the concentrations of these PCB congeners has decreased at a rate of approximately -0.00020 μg/L per year. (For details on the collection methods and laboratory techniques used in this study, refer to Jeremiason et al. (1994).) The researchers believe that, due to the remote location of Lake Superior and the absence of significant point source loadings, the decline in PCB concentrations represents a continental decrease in atmospheric loading of PCBs. (Other research suggests that cycling of PCBs in the environment, including volatilization of gaseous PCBs from waterbodies, is an important consideration (see Chapter IV).) According to the data, the concentrations of PCBs in Lake Superior were above the AWQC for human health until approximately 1983 and still remain above the GLWQC (criteria shown in Table II-2).

TABLE II-3
Concentration of Total PCBs in Lake Superior Water Column

Year	Total PCB Concentration (µg/L)	Total Concentration of 25 PCB Congeners (µg/L)
1978	0.00173 ± 0.00065	NA
1979	0.00404 ± 0.00056	NA
1980	0.00113 ± 0.00011	0.00099 ± 0.00010
1983	0.0008 ± 0.00007	0.00073 ± 0.00006
1986	0.00056 ± 0.00016	0.00055 ± 0.00015
1988	0.00033 ± 0.00004	0.00020 ± 0.00001
1990	0.00032 ± 0.00003	0.00021 ± 0.00001
1992	0.00018 ± 0.00002	0.00009 ± 0.00001

NA = Not applicable
Source: Jeremiason et al. 1994.

FISHING RESTRICTIONS AND FISH CONSUMPTION ADVISORIES

Another measure of contamination of the Great Waters caused by selected pollutants of concern is the existence of fishing restrictions or fish consumption advisories. These advisories are established as a means of limiting human exposure when fish taken from a particular body of water are found to contain levels of pollutants that exceed recommended intake levels (see sidebar on next page). Such advisories have immediate significance to the general public by providing concrete examples of health concerns and affecting the public use of waters and aquatic resources. States issue several different types of advisories for waterbodies in an effort to reduce health risks associated with exposure to pollutants in certain freshwater fish and shellfish species:

- *Informational health advisories*: advisories that fish tissue contains contaminants but not at levels high enough to warrant advising people to limit consumption.

- *Advisories to limit fish consumption*: advisories to either the general population or subopulations potentially at greater risk (e.g., pregnant or nursing women, those who fish for subsistence reasons) to restrict the size and frequency of meals of fish and shellfish;

- *Advisories against fish consumption*: advisories to either the general population or subpopulations potentially at greater risk (e.g., pregnant or nursing women) against consuming fish and shellfish;

- *No-kill zones*: notification that it is illegal to take, kill, or possess any fish from the specified waters; and

- *Commercial fishing bans*: bans on the commercial harvest and sale of fish and shellfish from the specified waterbody.

Interpreting Fish Advisory Data

Individual states are responsible for issuing fish advisories. Generally, an advisory is issued for a particular waterbody (or portion of waterbody), pollutant, fish species, and advisory type. In many advisories, the size of the fish affects the type of advisory issued (e.g., for walleye < 22", restricting meals in the general population may be advised, while for walleye > 22" not consuming the fish may be advised). For several reasons, comparing advisories quantitatively (e.g., counting the number of advisories per waterbody) is difficult and therefore, this Report does not do so. For example, a waterbody may appear to have more fish advisories than another waterbody, but it may be that: (1) more states are involved (e.g., advisories in Lake Ontario are issued only by New York, while four states issue advisories for Lake Michigan; (2) states have different methods or use different standards for identifying fish species affected by advisories, some of which may be more comprehensive than others; or (3) a state may issue an advisory for "all fish" making it difficult to count this advisory with advisories for particular fish species.

State advisory data are collected by EPA in a national data base. For each advisory, the data base contains information such as waterbody name, pollutant name, fish species, population targeted by advisory (called advisory type in this report), advisory status (e.g., active), and a contact name and telephone number. The data base does not, however, contain information on the levels of pollutants in fish or the benchmark levels set by a particular state for each advisory type. The information in the data base available for use in this report was current through 1995 (U.S. EPA 1996b). For this report, EPA reviewed the data base for any of the above advisories related to the Great Waters pollutants of concern in the Great Lakes, Lake Champlain, Chesapeake Bay, and several coastal waters.[7]

According to the fish advisory data base, no informational health advisories were in effect in any of the Great Waters. A portion of the Hudson River, which empties into the New York/New Jersey Harbor, had an active no-kill zone for all fish related to PCBs. Table II-4

[7] A more detailed description of the criteria EPA used to obtain information related to the Great Waters from the fish advisory data base is provided in Appendix B.

lists the commercial fishing bans that were in effect in the Great Waters. As the table shows, the commercial fishing bans in the Great Waters are all due to PCBs or dioxins.

TABLE II-4
Commercial Fishing Bans in the Great Waters

Waterbody (State Issuing Advisory)	Great Waters Pollutant of Concern	Fish Species
Lake Champlain (NY)	PCBs	Yellow perch
Lake Ontario (NY)	Dioxins, PCBs	Eel
Long Island Sound (NY)	PCBs	Striped bass
New York/New Jersey Harbor (various waters; NJ, NY)	PCBs	American eel, blue crab, carp, goldfish, striped bass
	Dioxins	Blue crab, crustaceans, fish, shellfish, striped bass

Source: U.S. EPA 1996b.

There are many active advisories to limit or avoid fish consumption in the Great Waters. Table II-5 (the Great Lakes and Lake Champlain) and Table II-6 (selected coastal waters, including Chesapeake Bay) indicate the type of advisories in effect for the Great Waters pollutants of concern, in increasing order of severity of the advisory (i.e., from advisories to at-risk subpopulations to restrict fish consumption up to advisories to the general population to not eat certain fish). For the Great Lakes and Lake Champlain, only lakewide advisories are included in Table II-5. Advisories for particular "hot spots" in these lakes (e.g., Green Bay), as well as the lakewide advisories, are presented in Appendix B. In Table II-6, the advisories for coastal waters represent the advisories as they are identified in the data base. Therefore, advisories for some coastal waters represent the entire waterbody (e.g., Tampa Bay), while others represent smaller estuaries or portions of the waterbody (e.g., Baltimore Harbor). Appendix B provides details on the waterbodies that represent the coastal water advisories. The fish consumption advisories shown in the tables have been issued for at least one fish species and, in many cases, have been issued for several fish species. For more detail on the fish species affected by the advisories and the states that issued the advisories, refer to Appendix B.

As shown in Table II-5, fish consumption advisories in the lakes of the Great Waters are most commonly associated with PCBs, followed by mercury, dioxins, and chlordane. For one lake, Lake Superior, an advisory related to toxaphene also has been issued. In the selected coastal waters, as shown in Table II-6, fish consumption advisories are most commonly associated with PCBs, followed by dioxins. Several advisories related to mercury, chlordane, and DDT also have been issued. Current fish advisories generally are associated with the same pollutants of concern as in the First Report. Furthermore, in this report, fish advisory data are available in more detail than were reported in the First Report (e.g., advisories by each state), especially because of the recent availability of the fish advisory data base. In addition, some states may have become more aggressive in their fish advisory programs. Therefore, the increase in number of advisories presented in Appendix B does not necessarily reflect a higher level of contamination in the Great Waters.

TABLE II-5
Fish Consumption Advisories in the Great Lakes and Lake Champlain

Pollutants of Concern	Great Lakes						Lake Champlain
	Lake Superior	Lake Michigan	Lake Huron	Lake Erie	Lake Ontario	Connecting Channels	
Chlordane	■	● □ ■	● □ ■	■			
Dioxins			● □ ■		● □ ■	● □ ■	
Mercury	○ ● □ ■	○ ●				○ ● ■	● □
PCBs	● □ ■	● □ ■	● □ ■	● ■	● □ ■	● □ ■	● □
Toxaphene	■						

TABLE II-6
Fish Consumption Advisories in Selected Coastal Waters

Pollutants of Concern	Chesapeake Bay	Other Coastal Waters					
		Long Island Sound	New York/New Jersey Harbor	Delaware Bay	Tampa Bay	Galveston Bay	San Francisco Bay
Chlordane	●	●		■	●		
Dioxins			○ ● □ ■			● □	○ ● □ ■
DDT							○ ● □ ■
Mercury							○ ● □ ■
PCBs	●	● □ ■	● □ ■	○ ● ■	●		○ ● □ ■

KEY: ○ Advisories to subpopulations potentially at greater risk (e.g., pregnant women) to restrict the size and frequency of meals of fish and shellfish
● Advisories to the general population to restrict the size and frequency of meals of fish and shellfish
□ Advisories to subpopulations potentially at greater risk (e.g., pregnant or nursing women) against consuming fish and shellfish
■ Advisories to the general population against consuming fish and shellfish

Source: U.S. EPA 1996b.

II.B Contamination of Biota

Measurements of pollutant levels in biota provide information about the extent of contamination in the waterbody, as well as potential bioaccumulation in the food web. Section 112(m)(1)(E) of the CAA requires EPA to sample biota, including fish and wildlife, in the Great Waters for hazardous air pollutants and to identify the sources of these pollutants. Because studies are already being performed under national programs such as the National Status and Trends Program, Great Lakes National Program Office, Chesapeake Bay Program, as well as state programs, EPA relies largely on these studies to support implementation of this CAA requirement. These programs provide information on the extent of contamination in the waterbodies, as reflected in tissues of living organisms. Contamination of biota also suggests potential contribution from various exposure routes such as air and water, as well as bioaccumulation in the food web. This section presents a brief overview of biota sampling approaches and how the sampling data provide useful information for assessing the extent of pollutant contamination in a waterbody. This overview includes a summary of two large-scale studies that are designed to assess national pollutant levels in aquatic biota, and how the results from these studies can apply to the Great Waters. This discussion is followed by a summary of research efforts addressing biota contamination specific to the Great Waters.

Sampling Biota for Contamination

Different sampling approaches are used to determine pollutant levels in biota because each monitoring study has its own objectives, such as to identify "hot spots" or characterize a waterbody's general condition. Therefore, caution must be taken in the interpretation of data, as well as statistical analyses applied to these data. To estimate spatial or temporal patterns of contamination, sophisticated sampling designs are often used. Figure II-1 illustrates the importance of sampling various components of an aquatic ecosystem when attempting to characterize the condition of the system.

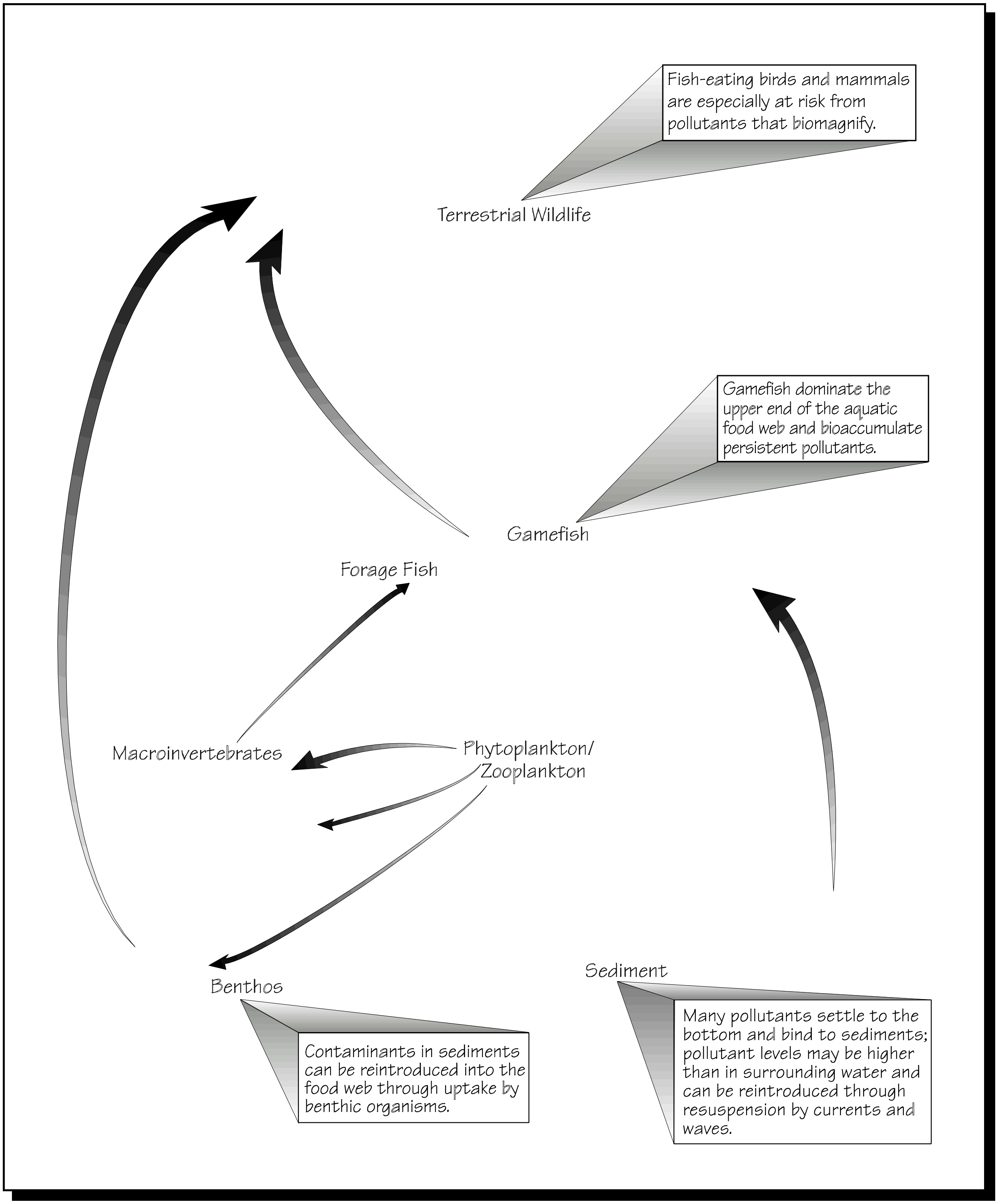
An Example of Bioaccumulation in the Food Web

Of the pathways by which ecosystems and components of ecosystems may be exposed to atmospheric mercury, exposure of high trophic level predatory wildlife to mercury in food is particularly important. Mercury biomagnifies in aquatic food chains, with the result that mercury concentrations in tissue increase as trophic levels increase. Therefore, the trophic level and feeding habits of an animal influence the degree to which that species is exposed to mercury. Predatory animals primarily associated with aquatic food chains accumulate more mercury than those associated with terrestrial food chains. Thus, fish eaters and their predators generally have the highest exposure to mercury. Species with high tissue levels of mercury include otter and mink, which are top mammalian predators of aquatic food chains. Top avian predators of aquatic-based food chains include raptors such as the osprey and bald eagle.

Sources: Bloom 1992; Eisler 1987a; Roelke et al. 1997; Wobeser and Swift 1976; Wren 1985.

In assessing contamination in a waterbody, aquatic and terrestrial species are often collected and analyzed. Game fish are useful bioindicators because they have long life spans, dominate the upper end of the aquatic food web, and can bioaccumulate many of the persistent pollutants (see sidebar). Their population levels may be affected by continued exposure to environmental stresses, such as eutrophication and pollutant contamination, and people become aware of changes in abundance of game fish. Reduced dissolved oxygen concentrations in the water may affect growth, survival, or structural development of fish,

FIGURE II-1
Assessing Contamination in a Waterbody



while pollutant contamination may result in decreased growth, reproduction, or survival of fish (see Section II.C for information on ecological effects). Contamination in fish can enter the diet of humans and other animals (see sidebar). Therefore, terrestrial wildlife, such as fish-eating birds or mammals, are often monitored. While all animals in the aquatic environment have the potential to be affected by pollutants, fish-eating birds and mammals are especially at risk from pollutants that biomagnify because they are frequently exposed to high levels of these pollutants.

Contaminated sediments pose a potential hazard to human health due to biomagnification up the food web. Many pollutants in the water settle to the bottom and bind to sediments or remain in solution in the water between the sediment particles. Although some pollutants can be degraded by bacteria, many persist in sediments for years, even after the original source of contamination has been removed (Howdeshell and Hites 1996). Therefore, pollutant levels in the sediment may be higher than the concentrations in the surrounding water and/or biota, and should be interpreted cautiously when extrapolating to potential levels in biota. Pollutants in the contaminated sediment can also be reintroduced into the water column and can enter the food web through benthic (bottom-dwelling) organisms (e.g., clams, crustaceans, and worms), which are prey to larger fish. Benthic community structure, as well as pollutants in tissues of benthic organisms, may serve as useful indicators of sediment contamination.

EPA Guidance For Issuing Fish and Wildlife Consumption Advisories

In response to a 1990 request from the states, EPA established The Fish and Wildlife Contamination Program to assist states and tribes assess and reduce health risks associated with exposure to chemical contaminants in noncommercially harvested (e.g., subsistence) fish and wildlife. In partnership with states and tribes, EPA has developed a series of guidance documents which provide a scientifically sound, cost-effective method for developing, issuing, managing, and communicating fish consumption advisories. Though many states use the EPA guidance for developing fish and wildlife consumption advisories, some states use parts of the guidance selectively. A few states still apply outdates methods for characterizing contaminants. EPA continues to work with the states and tribes to establish a national consistency in the methods used for characterizing the risks posed by contaminants in noncommercial fish and wildlife.

The Fish and Wildlife Contamination Program has also worked with the states and tribes in organizing training workshops and national conferences. In addition, EPA manages national data bases such as the *Listing of Fish and Wildlife Consumption Advisories* (discussed in Section II.A). The listing, which is updated annually, includes an inventory of all fish and wildlife consumption advisories issued by the states and tribes. Because of additional sampling due to the increased attention given to fish contamination, the number of advisories issued by the states and tribes has increased 72 percent, from 1,278 advisories in 1993 to 2,193 advisories issued in 1996. Most of these advisories have been issued due to mercury contamination.

Wide-scale monitoring studies have been conducted on pollutant contamination of biota to assess large-scale regional and national impacts in U.S. waters. These programs provide valuable information about major pollutants (e.g., where they are found and where they bioaccumulate) and the extent of contamination in the waterbodies, although the source of contamination (or contribution of atmospheric deposition) is generally not evaluated.

Two large-scale programs that assess the contamination of biota in major U.S. waterbodies or coastal areas are the NOAA National Status and Trends (NS&T) Program and the EPA Environmental Monitoring and Assessment Program (EMAP). The NS&T Program monitors trends of more than 70 chemical contaminants (organic compounds and trace metals) in bottom-feeding fish, shellfish, and sediments at almost 300 coastal and estuarine locations throughout the United States. A well-known project of the NS&T is the Mussel Watch. EMAP has carried out regional-scale studies, on both terrestrial and aquatic species. The data collected for EMAP-Estuaries, to quantify conditions in coastal regions of the United States, are useful to the Great Waters. Sampling and subsequent analyses in EMAP studies are generally not directed at a specific waterbody, but encompass a larger geographic region, which may include waterbodies of the Great Waters. These two projects provide information on assessing the extent of pollutant contamination in U.S. waterbodies; however, the application of their findings to individual waterbodies is limited since these projects were not designed to address concerns specific to the Great Waters.

NS&T MUSSEL WATCH PROJECT

Initiated in 1986, this continuing project is directed at tracking temporal trends in concentrations of pollutants (e.g., PAHs, PCBs, pesticides, metals) found in whole soft-parts of mollusks at about 255 coastal and estuarine sites on the Atlantic, Pacific, and Gulf coasts (including Great Waters such as Chesapeake Bay and Galveston Bay).

> **Systematic Sampling Approach**
>
> The Mussel Watch project is designed to provide long-term and large-scale monitoring of pollutant distribution, looking for temporal (not spatial) trends. The sampling design is set up to be *representative of large areas* rather than small-scale patches of contamination. The objective is to get a representative picture of the general or "average" conditions of the U.S. coastal waters. Therefore, a systematic sampling approach is used. Also, it is useful for estimating statistically the average concentration of pollutants when general trends or patterns in concentration are known from other sources of information.

Since no single species of mollusk is common to all waters, a number of species have been sampled for this project, including several common to the Great Waters — blue mussels (*Mytilus edulis*) from the Northeast and West Coasts, American oysters (*Crassostrea virginica*) from the Mid-Atlantic and Gulf coasts, and zebra mussels (*Dreissena polymorpha*) from the Great Lakes. Mussels and oysters are useful for monitoring changes in the pollutant levels because they remain at fixed sites, and concentrations in their tissues reflect, in general, changes in the concentrations in the surrounding water. There are species differences, so data need careful interpretation. Sampling for this project is performed during the same season each year to reduce the influence of seasonal cycles on natural factors (e.g., salinity, reproductive state). Sites that were selected support adequate populations of these mollusks such that sufficient samples are available annually over many years (O'Connor 1992; O'Connor and Beliaeff 1995).

Table II-7 is a general representation of the pollutant trends in mollusks during the 1986-1993 period at Great Waters sites based on analysis of data collected for the Mussel Watch Project (O'Connor and Beliaeff 1995). All of the trends shown in Table II-7 are statistically significant (confidence level of 90 percent and above); however, quantitative information regarding the changes in pollutant levels was not provided by O'Connor and Beliaeff (1995). Consequently, the magnitudes of the increases or decreases cannot be compared between sites (i.e., one cannot determine which sites showed the most improvement). However, useful information regarding general pollutant trends is presented.

As shown in Table II-7, most of the Great Waters sites did not exhibit a statistically significant trend or change in pollutant levels in mollusk tissues during the eight-year period.

Trends, when present, were mainly downward since 1986 (i.e., for cadmium, PCBs, DDT, PAHs, chlordane, and dieldrin). According to the investigators, decreases in levels of these chemicals are probably the result of bans on the use of chlorinated hydrocarbons and the reduced use of certain pollutants (O'Connor and Beliaeff 1995).

The level of mercury in mollusks showed an increasing trend over the eight-year period only in one Galveston Bay site (O'Connor and Beliaeff 1995). During the four-year period from 1986 to 1990, mercury concentrations increased at several other sites (O'Connor 1992); however, mercury levels stabilized at these sites by 1993 so that no statistically significant trend was apparent over the eight-year period (O'Connor and Beliaeff 1995). Lead levels in mollusks also exhibited an increasing trend at one site in Galveston Bay and one site in Tampa Bay during the eight-year period (O'Connor and Beliaeff 1995); however, no change in lead levels was evident at these sites between 1986 and 1990 (O'Connor 1992). The investigators concluded that because the high concentrations of mercury and lead were found, for the most part, in the vicinity of population centers, they may be attributable to human activities (O'Connor and Beliaeff 1995).

The information presented in Table II-7 and discussed above focuses only on trends in contaminant levels in mollusks based on the Mussel Watch Project data. For the pollutants that show no significant trends over the eight-year period, it does not necessarily mean that no changes are occurring with respect to contamination of biota at Great Waters sites. Other analyses of the Mussel Watch Project data may indicate different patterns of pollutant concentrations in mollusks. In addition, gamefish and fish-eating birds and mammals are much higher in the food web than mussels and oysters, and therefore, may exhibit different patterns of pollutant concentrations in their tissues.

TABLE II-7
Eight-Year Trends of Pollutant Concentrations in Mussel Watch Project (1986-1993)

Waterbodies (number of sites examined)	Contaminant Trend (number of sites affected)[a]							
	Mercury	Lead	Cadmium	PCB	DDT	PAH	Chlordane	Dieldrin
Chesapeake Bay (5 sites)	↔(5)	↔(5)	↓ (1) ↔(4)	↓ (3) ↔(2)	↓ (3) ↔(2)	↔(5)	↓ (4) ↔(1)	↓ (3) ↔(2)
Delaware Bay (4 sites)	↔(4)	↔(4)	↓ (1) ↔(3)	↓ (2) ↔(2)	↔(4)	↔(4)	↓ (1) ↔(3)	↔(4)
Long Island Sound (9 sites)	↔(9)	↓ (1) ↔(8)	↓ (4) ↔(5)	↓ (5) ↔(4)	↓ (2) ↔(7)	↓ (1) ↔(8)	↓ (3) ↔(6)	↓ (1) ↔(8)
Narragansett Bay (2 sites)	↔(2)	↓ (1) ↔(1)	↔(2)	↔(2)	↔(2)	↔(2)	↔(2)	↔(2)
Tampa Bay (3 sites)	↔(3)	↑ (1) ↔(2)	↔(3)	↓ (1) ↔(2)	↓ (1) ↔(2)	↔(3)	↓ (2) ↔(1)	↔(3)
Galveston Bay (6 sites)	↑ (1) ↔(5)	↑ (1) ↔(5)	↔(6)	↓ (2) ↔(4)	↓ (1) ↔(5)	↔(6)	↓ (4) ↔(2)	↓ (3) ↔(3)

[a] Represents trends in annually measured concentrations of contaminants in mollusks over 8-year period; trend indicated by arrow (i.e., ↓ = decreasing trend; ↑ = increasing trend; ↔ = no trend). Number of sites showing trend within each waterbody is indicated in parentheses. Sites were sampled in at least six of the eight years. All trends shown are statistically significant.

Adapted from O'Connor and Beliaeff (1995).

EMAP-ESTUARIES

> **EMAP** is a national program initiated in 1989 in response to the EPA Science Advisory Board's recommendation to monitor the status and trends of the U.S. ecological resources -- terrestrial, freshwater, and marine. The program is directed by EPA's Office of Research and Development, with participation by other federal agencies (e.g., NOAA, U.S. Forest Service, U.S. Fish and Wildlife Service).

One goal of EMAP is to quantitatively evaluate the condition of coastal estuaries, by investigating several environmental conditions: hypoxia (low oxygen levels), sediment contamination, coastal eutrophication, and habitat loss. A probability-based approach is used, which allows estimates to be made of the uncertainty associated with assessments and improves the ability to identify ecological responses to pollution. Of interest to the Great Waters are the EMAP-Estuaries results obtained for the Virginian Province (Cape Cod to the Chesapeake Bay) and Louisianian Province (Texas to west coast of Florida). Statistical data collected provide primarily quantitative information on a regional scale.

Results are available for the Virginian Province for 1992 (trend data for 1990-1993 are currently being analyzed), which include sampling results for two relevant Great Waters sites, the Chesapeake Bay (as well as connecting tributaries and small water systems) (53 sampling stations) and Long Island Sound (14 stations). Together, the two waterbodies represent approximately 63 percent of the surface area of the entire province. One of the environmental indicators assessed in the study was sediment contamination. Results showed that metal concentrations in the sediment for the Chesapeake Bay were similar to the concentrations for the overall Province ($\approx$24 μg/g for lead; $\approx$0.054 μg/g for mercury; $\approx$0.206 μg/g for cadmium). In Long Island Sound, the concentrations of some metals in the sediment (44.2 μg/g lead; 0.088 μg/g for mercury) were higher than those reported for the Chesapeake Bay and the overall Virginian Province. These results suggest that Long Island Sound exhibits slightly higher contamination of some metals in sediment compared to most other waterbodies in the east coast of the United States. Comparing results among specific waterbodies within the regional area is limited because the density of sampling points was not designed to thoroughly characterize each waterbody separately. Also, uncertainty exists in the analysis of these waterbodies due to the short data collection period (one year) (Strobel et al. 1994).

Biota Contamination by Major Waterbody

This subsection presents information from some monitoring studies and investigations of the pollutant levels in tissues of biota living in the Great Waters. These studies, most of which measure concentrations of metals and organochlorines in fish, provide evidence that toxic contamination is occurring in biota from these waters. In most instances, the researchers focused on pollutant levels in tissues of the biota, and did not look for toxic lesions or other adverse effects. Nevertheless, some of the long-term studies provide information on potential contamination trends in the species examined. In addition, studies that monitor toxic contamination in sediment are presented because contaminated sediment may be a long-term source of pollutant exposure to the aquatic biota and food web. These studies suggest continuing contamination in the waterbody.

GREAT LAKES

Contaminant concentrations in gamefish from the open waters of the Great Lakes have been monitored for over 20 years and provide one of the most extensive data bases on trends of environmental contaminants in organisms at the upper end of the food web. Three monitoring efforts that data have come from include: (1) lake trout monitored by Fisheries and Oceans Canada; (2) lake trout and walleye cooperatively monitored in U.S. waters by EPA's Great Lakes National Program Office, U.S. National Biological Service, and the Great Lakes States; and (3) coho salmon fillets cooperatively monitored in U.S. waters by the Great Lakes States, FDA, and EPA's Great Lakes National Program Office.

PCB Contamination in Great Lakes Biota

Lake Trout/Walleye: During the period 1977-1992, PCB concentrations in lake trout, as well as walleye in Lake Erie, declined significantly, but in recent years, concentrations have generally remained stable or increased slightly in Lakes Michigan, Huron, Superior, and Erie.

Coho Salmon: PCB concentrations in coho salmon collected from Lake Michigan declined from 1.9 μg/g (1980) to 0.38 μg/g (1983), but then increased to 1.09 μg/g (1992). A similar pattern was observed in Lake Erie and the upper reaches of the Saint Lawrence River.

Herring Gull Eggs: Monitored since 1974, the greatest decline in PCB contamination in herring gull eggs occurred between 1974 and 1981. Since then, the rate of decrease has leveled off, and by 1991, slight increases were reported in the levels of some PCBs.

Sources: De Vault et al. 1995, 1996.

These monitoring efforts have demonstrated that, while significant declines in PCB and DDT concentrations in lake trout, walleye, and coho salmon have been observed over the past two decades, the amount of residues of PCBs and DDT in these fish have leveled off or even increased slightly in the last ten years (De Vault et al. 1995, 1996) (see sidebar). This change in trend has occurred despite declining ambient water concentrations of PCBs. A similar trend has been noted in the levels of PCBs and DDT in herring gull eggs, which have been monitored by the Canadian Wildlife Service since 1974.

Lake condition changes can complicate pollutant contamination issues. The strong correlation between trends in DDT and PCBs suggests that changes in the composition of the food web (or trophic structure) may be partly responsible for increases in contaminant concentrations at the upper end of the food web (i.e., gamefish) (De Vault et al. 1995, 1996). For example, research by Haffner (1994) and Stow et al. (1995) on the exposure dynamics of organic pollutants in Lakes Erie and Michigan suggests that changes in the food web could be the cause of the observed increase in PCB contamination in biota. Large, regional pools of PCBs can be reintroduced from sediments by benthic organisms. Changes in the species composition at the mid-trophic levels of the food web may biomagnify greater amounts of PCBs to higher trophic levels. Increased PCB levels in certain predator fish also may be due to reductions in their growth rate.

Evidence of changes in the exposure dynamics of organic contaminants has been observed in the western basin of Lake Erie with the invasion of zebra and quagga mussels (Haffner 1994). These mussels increase biomagnification of pollutants in the benthic food web by consuming significant amounts of phytoplankton that are contaminated with pollutants. The major predator of the zebra mussel is the drum (a low-trophic level fish), which in turn is a preferred prey of the herring gull. By concentrating contaminants contained in phytoplankton and other suspended organic particles, zebra and quagga mussels can cause PCB levels in certain fish and in herring gulls to increase even though ambient water concentrations of contaminants are decreasing.

> **Sediment Core Data in the Great Lakes**
>
> Atmospheric pollutant loadings into the Great Lakes region are estimated from mass balance studies and modeling data, although indirect measures of contaminant loadings, such as sediment core data, are also desirable. Recent measurements of sediment core data have shown declining concentrations of PAHs in Lake Michigan, PCBs and DDT in Lakes Michigan and Ontario, and lead and mercury in Lakes Superior, Michigan, and Ontario. Comparison of sediment data between the Great Lakes provides additional information on sources of loadings. For example, toxaphene has long been thought to result from long-range atmospheric transport from the southeastern U.S.; however, sediment cores from Lake Superior and upper Lake Michigan suggest little decline in toxaphene (contrary to declines observed for DDT, mercury, and PCBs in these lakes). Efforts are underway to examine this issue.
>
> Sources: De Vault et al. 1995; Simcik et al. 1996.

The three monitoring programs discussed above also provide information on the levels of two pesticides, dieldrin and toxaphene, in upper trophic-level fish from the Great Lakes. Dieldrin concentrations have exhibited a general pattern of decline in the Great Lakes since the 1970s (De Vault et al. 1995, 1996). Peak levels were noted in 1979 in Lakes Michigan, Superior, Huron, and Ontario and again in 1984 in Lakes Superior, Huron, Ontario, and Erie. Toxaphene concentrations are highest in lake trout from Lakes Michigan and Superior and lowest in lake trout from Lakes Erie and Ontario (De Vault et al. 1996). Lake trout from Lake Michigan sampled between 1982 and 1992 suggest declining levels of toxaphene during this period; however, there was no significant change in concentrations of Lake Superior lake trout (Glassmeyer et al. 1997). A similar trend was observed in rainbow smelt. The drop in toxaphene concentrations coincides with the U.S. ban on toxaphene in 1982. The investigators speculate that the lack of decline in Lake Superior is due to either a lake-specific source that continues to load toxaphene into Lake Superior or a slower removal rate in Lake Superior compared to the other Great Lakes (more discussion in Section IV.A on the Great Lakes).

In contrast to the monitoring studies of gamefish, monitoring of forage fish provides an indication of contamination at lower levels of the food web. Rainbow smelt have been routinely monitored in Lakes Superior, Huron, Erie, and Ontario by Fisheries and Oceans Canada since 1977. During this time, concentrations of PCBs, mercury, and total DDT have declined significantly in smelt from these lakes (De Vault et al. 1995). Smelt from Lake Ontario consistently have the highest tissue concentrations of PCBs (~0.5–2.25 μg/g) and total DDT (~0.15–0.6 μg/g), while those from Lake Superior have the highest mercury levels (~0.02–0.1 μg/g).

Contaminant concentrations in young-of-the-year spottail shiners are useful indicators of local, recent pollutant inputs into aquatic ecosystems because they do not travel extensively during their first year of life. Surveillance of these fish by the Ontario Ministry of Environment and Energy (primarily in Canadian waters) has shown a general decline in tissue PCB and DDT concentrations (De Vault et al. 1995). Contaminant levels also have been assessed in young-of-the-year fish from the New York waters of the Great Lakes (Skinner et al. 1994). Elevated concentrations of PCBs were found mainly in fish from the St. Lawrence River drainage area below the Moses-Saunders Dam in Massena; these levels were attributed to industrial activities in the area. Levels of mercury in these fish were low (<100 ng/g).

Zebra mussels have been considered to be potential system-wide biomonitors of organic contamination trends in the Great Lakes (Comba et al. 1996). Researchers collected and analyzed specimens from 24 sites in Lake Erie, Lake Ontario, and the St. Lawrence River between 1990 and 1992 for residues of PCBs and organochlorine pesticides. Mean concentrations of 154 ng/g total PCBs, 8.4 ng/g total DDT, and 3.5 ng/g total chlordane (whole mussel dry weight basis) were reported. Concentrations varied greatly between sites (e.g., 22–497 ng/g total PCBs), which the researchers attributed to the sensitivity of these mussels to different levels of contamination (Comba et al. 1996). The investigators also indicated that the observations of spatial contaminant trends in the study were similar to findings from other biomonitoring programs.

LAKE CHAMPLAIN

Current efforts to monitor toxic pollution in Lake Champlain have focused on fish and sediment contamination by metals and organic compounds. The Vermont Department of Environmental Conservation implemented a study to analyze soft tissue from mussels (*Elliptio complanata*) as a bioindicator for the lake. Mussels were collected at mouths of several Lake Champlain tributaries; chlordane and PAHs were detected in the mussels (LCBP 1994). In 1987-1988, the States of Vermont and New York analyzed fish tissue collected from Lake Champlain for 17 contaminants. Elevated levels of PCBs were found in large lake trout and in American eel and brown bullhead. The findings of this study led, in part, to health advisories being issued against eating these fish species in Lake Champlain (LCBP 1994).

Because of elevated levels of pollutants in Lake Champlain, the Lake Champlain Sediment Toxics Assessment Program was initiated (McIntosh 1994). Pollutants that were measured included trace metals (cadmium, mercury, lead) and organic compounds (PCBs, PAHs, dioxins/furans). Pollutant levels were measured at nine sites during 1991 and 1992. Findings after the end of the first phase (May 1993) provided little evidence of widespread high-level contamination (although high levels of PCBs and PAHs were measured in sediment near two dock sites). The study did find widely varying patterns of contamination. It appears that, at some sites, some pollutants had higher concentrations in the deeper layers of the sediments, with the upper layers of sediment showing less contamination, while other pollutants exhibited a reversal of this pattern (McIntosh 1994); however, the investigator did not provide an explanation for this deposition pattern. The local and/or regional source of the contamination is not known.

As part of the Lake Champlain Sediment Toxics Assessment Program, a biological assessment of the contaminated sediments was also performed (McIntosh 1994). Most of the year, lake trout do not inhabit bottom waters near the sediment-water interface. However, concerns exist for the mechanisms that may link lake trout to PCB-contaminated sediment. This issue was evaluated by looking at one possible link, the freshwater shrimp *Mysis relicta* (or mysids). Mysids are believed to be a major component in the Lake Champlain food web, and the high lipid content of these organisms make them potential accumulators of PCBs. Laboratory experiments demonstrated that exposure to PCB-contaminated sediment results in high levels of PCBs in the mysids (McIntosh 1994). However, there was no attempt to predict the potential of mysids to redistribute PCBs within the sediments in Lake Champlain.

CHESAPEAKE BAY

Adverse effects that could be related to pollutants accumulating in the tissues of organisms, such as reduced growth, reproduction effects, and tumor development, have been reported in aquatic organisms in a variety of habitats in the Bay from the 1980s to the early 1990s. The Chesapeake Bay Program has sponsored forums to assess contaminant levels in biota and to reach a consensus regarding the trends in the pollutants found in biota and in sediment. While significant declines in metal contamination of fish tissue have been observed over the past two decades, elevated metal concentrations have been measured in fish in specific, more industrialized areas of Chesapeake Bay (CBP 1994b).

Studies conducted from 1970 to 1992 show that chemical contamination has caused various effects to wildlife in the Chesapeake Bay during the 1970s and 1980s (CBP 1994b). In 1991, the *Status and Assessment of Chesapeake Bay Wildlife Contamination Forum* was held to critically review data on the effects of exposure and uptake of pollutants on Chesapeake Bay basin birds, mammals, reptiles, and amphibians. The committee concluded that there was little evidence to suggest pollutants were posing a serious *direct* hazard to birds in the early 1990s. Instead, it is more likely that indirect effects on wildlife habitats and food sources (e.g., excessive nutrients, suspended sediments, herbicides) have greater impacts on bird populations. The forum found that there were insufficient information available at the time to fully assess potential adverse impacts of chemical contamination on mammalian, reptile, and amphibian populations in the Chesapeake Bay basin (CBP 1994b). Due to more recent scientific information on some pollutants' mechanisms of action such as endocrine disruption (see Sections II.C and II.D), the conclusions of the 1991 forum may need to be reevaluated with more current data on pollutant contamination in Chesapeake Bay biota.

In 1993, the *Chesapeake Bay Finfish and Shellfish Tissue Contamination Critical Issues Forum* was held to address the following issues: (1) magnitude and extent of fish and shellfish contamination in the Chesapeake Bay and its basin; (2) impact (i.e., bioaccumulation, toxicity) of the contamination at basinwide, baywide, regional, or local scales; and (3) comparison of the contamination to that of other waterbodies (e.g., Puget Sound, Great Lakes) (CBP 1994b). The data compiled by the forum indicate that finfish and shellfish tissue contaminant concentrations declined significantly after the 1970s for several metals, pesticides, and organic chemicals. For fish species combined, concentrations of PCBs and DDT in fish liver tissue are in the low range relative to national data. However, at the species-specific level, Atlantic croaker and spot collected from 1984 to 1987 had levels of chlordane, PCBs, dieldrin, and total DDT in the liver above the national average and national median for these species. Lead and mercury concentrations in croaker livers were generally above the national average and national median, while lead concentrations in spot livers were sporadically high. The concentrations of PCBs, chlordane, dieldrin, DDT, cadmium, and mercury in oysters in the Chesapeake Bay have declined between 1986 and 1991. Levels of mercury, chlordane, toxaphene, and PCBs in finfish from the Chesapeake Bay basin "hot spots" (e.g., Baltimore Harbor, Back River) are generally well below those found at other areas considered contaminated (e.g., New York/New Jersey Harbor, Lake Michigan, Boston Harbor).

OTHER COASTAL AREAS

Several monitoring studies of biota contamination also have been performed in other coastal waterbodies of the Great Waters. Some relevant findings are highlighted below;

however, additional research is needed to provide a more complete picture of biota contamination in some of these coastal areas.

- *Galveston Bay*. There is little information about historical trends and concentrations of pollutants in aquatic organisms from Galveston Bay. For this reason, the Galveston Bay National Estuary Program initiated a study to characterize pollutant contamination in edible fish and shellfish in the bay. Between 1991 and 1993, 14 fish species, two shellfish species, and three bird species were sampled for numerous pollutants, including several Great Waters pollutants of concern. No "hot spots" of biota contamination were detected and the fish tissue concentrations rarely exceeded FDA criteria for these contaminants (Brooks et al. 1992). The study did not evaluate dioxins, but fish consumption advisories to protect the general population currently exist for dioxins in Galveston Bay.

- *Tampa Bay*. Tissue concentrations of organic compounds and trace metals in Tampa Bay oysters (*C. Virginica*) (sampled between 1986 and 1989) were compared to reported levels for adverse effects to mussels at other sites (Long et al. 1991). The average concentration of total PCBs in the Tampa Bay oysters collected over four years was 0.15 ppm dry weight; total PCB concentration of 0.18 ppm or greater occurred in 11 of 55 samples. Investigators determined that, for total PCBs, adverse effects (e.g., reproductive effects, cellular damage, and biochemical changes) in mussels were associated with tissue concentrations of 0.18 to 1.43 ppm dry weight. The average concentration of mercury in the Tampa Bay oysters collected over four years was 0.27 ppm dry weight; mercury concentration of 0.4 ppm or greater occurred in 13 of 55 samples. Adverse effects (e.g., pathological and enzymatic responses) in mussels were associated with mercury tissue concentration of 0.4 ppm dry weight. Based on these data, Long et al. (1991) concluded that the PCB and mercury concentrations in some oysters in Tampa Bay may be sufficient to cause potential adverse biological effects.

- *New York-New Jersey Harbor and New York Bight*. In 1993, 23 species of fish, six species of bivalves, two species of crustaceans, and one species of cephalopod were collected from six areas of the New York-New Jersey Harbor, including the New York Bight Apex (Skinner et al. 1996). The samples were analyzed for contaminants, including PCBs, organochlorine pesticides, and mercury. Of these compounds, PCBs were the primary chemical contaminants of concern. Average total PCB concentrations for American eel, striped bass, white perch, bluefish, rainbow smelt, and the hepatopancreas of blue crab and American lobster in one or more areas of the harbor estuary exceeded the FDA tolerance level for PCBs (2,000 ng/g). In blue crab and American lobster, PCB and organochlorine pesticide residue concentrations were particularly elevated in tissues with high lipid content (e.g., hepatopancreas). The researchers noted that although relatively low levels of contamination were found in muscle tissue, increased contaminant levels in the hepatopancreas may present a substantially increased risk to those people who choose to eat this tissue (Skinner et al. 1996). PCB concentrations were highest in the Hudson River and Upper Bay, the East River, and the Arthur Kill-Kill Van Kull-Newark complex; concentrations were lowest in Jamaica Bay and the New York Bight Apex. The principal components of the PCB concentrations observed were the higher chlorinated congeners.

Residues of certain other contaminants (e.g., DDT, chlordane, dieldrin, and mercury) also exceeded regulatory criteria in some fish tissue samples (Skinner et al. 1996). Analyses were performed for hexachlorobenzene and toxaphene, but they were seldom or never detected.

II.C Ecological Effects

This section is intended to provide a brief overview of the current literature on the potential ecological effects (generally, adverse effects to aquatic organisms, birds, and mammals) from exposure to the Great Waters pollutants of concern. In general, the information presented in this section represents data published since the background document from the First Report to Congress on exposure and effects (Swain et al. 1992a) was completed through 1995. Because the information presented in this report covers only recent studies, it cannot be used alone to determine whether these effects are widespread in the environment. As in the First Report to Congress, the contribution of atmospheric deposition to toxic contamination and potential ecological effects associated with exposure to the pollutants cannot be quantified at this time.

This section presents information on effects observed in both laboratory and field studies; it does not, however attempt to establish a relationship between the two types of studies. This section also does not provide information on the exposure levels of the pollutants responsible for the observed effects. In addition, this section does not distinguish effects that may occur in wildlife with long-term exposure to the pollutants of concern from effects caused by acute, high-level exposures (e.g., accidental spills). The potential effects of a pollutant may vary with duration of exposure, possibly due to a breakdown of the chemical in the body to another chemical that is more toxic or affects other target organs than the exposed chemical. Furthermore, adverse effects on ecological health caused by exposure to toxic contaminants are not often easy to distinguish from other stresses. For example, fish populations in the Great Lakes suffer from habitat loss, overfishing, and the introduction of non-native species, in addition to the effects from toxic contaminant exposure (U.S. EPA 1995a). Finally, studies on ecological effects generally do not determine the exposure pathway of the pollutants (e.g., atmospheric deposition). In this report, under future directions for research, EPA recommends coordinated analyses of persistent pollutants that relate field measurements of concentrations to estimated exposure and associated effects observed in biota.

Conclusions from the First Report to Congress

Information from the First Report to Congress is presented here to provide a foundation for the subsequent discussion of the recent information available for this report. The First Report to Congress, as well as the background document on exposure and effects (Swain et al. 1992a), identified many adverse ecological effects, at both the individual species level and the ecosystem level, associated with the pollutants of concern. In addition, the First Report to Congress discussed eutrophication of estuarine waters, which is the main ecological effect relevant to this report associated with nitrogen loading. Research findings and studies presented in the First Report to Congress led to the following general conclusions concerning potential ecological effects:

- ♦ The selected pollutants of concern have been linked to a broad range of effects at the individual species level in aquatic organisms and other wildlife, including effects on the reproductive, nervous, immune, and endocrine systems, and changes in enzyme functioning.

- ♦ Reproductive effects of certain pollutants of concern include reduced fertility, increased embryo toxicity, reduced hatchability, reduced survival of offspring, abnormalities in

> **Pollutants of Concern for Reproduction**
>
> Pollutants of concern that have been linked with reproductive impairment in aquatic and terrestrial wildlife include cadmium, DDT/DDE, dieldrin, lead, lindane, mercury, PCBs, and 2,3,7,8-TCDD.

offspring, parental behavior change, and changes in mating behavior (e.g., impaired hormone activity, changed adult sexual behavior). For example, eggshell thinning in a number of bird species and associated reproductive loss were linked to exposure to DDT (and its metabolite DDE) in the 1960s and 1970s. Recent decreases in environmental concentrations of reproductive pollutants of concern, such as DDT and PCBs, are correlated with population recoveries in many bird and other wildlife species; however, some populations in certain regions of the Great Lakes still exhibit higher rates of reproductive failure than in other areas.

♦ Effects on the nervous and endocrine systems may occur at very low exposure levels. For example, wild populations of Great Lakes herring gulls, Forster's terns, and ring-billed gulls exposed to various pollutants of concern have exhibited behavioral changes such as female-female pairings, which result in abnormal incubation activities and nesting behavior, including nest abandonment.

♦ Several of the pollutants of concern cause changes in enzyme functioning. Studies reported that the activity of enzymes responsible for the breakdown of foreign compounds is greatly increased by most of the pollutants of concern. In fish, the increased activity of these enzymes has been shown to result from exposure to PCBs and PAHs. In birds, "wasting" syndrome (i.e., the condition in which an animal slowly loses body weight until it can no longer sustain itself) has been related to altered enzyme activity resulting from exposure to pollutants such as TCDD.

♦ Exposure of communities of bottom-dwelling aquatic species in the Great Lakes to toxic chemicals has resulted in significant changes in species diversity and populations. In addition, fish-eating birds such as bald eagles, herring gulls, and Forster's terns in the Great Lakes region have undergone significant population declines since the 1960s. Only in recent years, as water concentrations of pollutants in the Great Lakes have declined, have some species begun to recover. Certain current population recoveries of fish-eating birds are still dependent on migration to Great Lakes breeding colonies from other areas.

♦ Eutrophication[8] is one of the most serious pollution problems facing estuarine waters of the United States. Atmospheric deposition of various nitrogen compounds (mostly nitrates and ammonium) can contribute significantly to eutrophication in coastal waters where productivity is usually limited by nitrogen availability. Accelerated eutrophication results in severe ecological effects such as nuisance algal blooms, dieback of underwater plants (due to reduced light penetration), reduced oxygen levels in the water, and reduced populations of fish and shellfish. The reduction in oxygen levels

[8] Eutrophication is an overabundance of nutrients (e.g., nitrogen) in a water body. It is a natural process that typically takes place over hundreds of years, but can be greatly accelerated by additions of nutrients from human activities.

may reduce or eliminate bottom-feeder populations, create conditions that favor different species, or cause dramatic fish kills, resulting in an altered food web.

The remainder of this section presents updated information on ecological effects associated with the Great Waters pollutants of concern.

Current Understanding of Ecological Effects

Since the First Report to Congress, updated information on the pollutants of concern and their effects on aquatic and terrestrial wildlife has become available. This section first discusses some notable research efforts on ecological effects relevant to some Great Waters pollutants of concern and then provides a brief overview of recent data on the potential ecological effects specific to each pollutant of concern.

As introduced in the First Report to Congress, the role of endocrine disruptors in causing adverse effects wildlife and humans is an emerging and controversial issue. Endocrine disruptors were termed "environmental estrogens" in the First Report to Congress; however, because the interference with hormone action was found not to be limited to estrogen, these pollutants are now more generally referred to as "endocrine disruptors." For example, p,p'-DDE (a breakdown product of DDT) has been shown to inhibit the binding of androgen, a male hormone, to receptors, among other androgen actions (Kelce et al. 1995). Several pollutants have been identified as possible endocrine disruptors, including 11 of the 15 pollutants of concern for atmospheric deposition to the Great Waters: chlordane, dieldrin, DDT/DDE, hexachlorobenzene, lead, lindane, mercury (in the form dimethylmercury), PCBs, TCDDs, TCDFs, and toxaphene (e.g., Cassidy et al. 1994; Chowdhury et al. 1993; Colborn et al. 1993; McKinney 1994; Soto et al. 1994; U.S. EPA 1994c). Since the First Report to Congress, scientific research on endocrine disruptors has continued to provide evidence of their adverse effects and has investigated their mechanisms of action (i.e., how they disrupt the endocrine system within the body).

The existence and effects of the hormone-like action of environmental pollutants were first hypothesized in the 1950s and 1960s. In the late 1980s, scientists concerned with noncancer effects of toxic pollutants brought this issue into focus. In July 1991, many scientists whose diverse research interests touched on some aspect of endocrine system disruption convened at the Wingspread Conference. The conference helped identify future research needs for improving the understanding of endocrine disruptors, their mechanisms of action, and their effects (NWF 1994). Many of the adverse effects in wildlife and humans (e.g., reproductive, developmental, and immunological effects) associated with the pollutants of concern are now theorized to be associated with the endocrine-disrupting action of the pollutants. Recent articles published in the mass media have brought this issue widespread attention (Begley and Glick 1994; Suplee 1996; Weiss 1994; Weiss and Lee 1996).

Endocrine disruptors are believed to interfere with the operation of the endocrine system in many ways, such as by mimicking natural hormones or by blocking natural hormones. This interference can potentially disrupt the reproductive and immune systems and adversely affect metabolism, growth, and behavior.

Some of the recent articles demonstrating endocrine disruption by a few of the Great Waters pollutants of concern in aquatic and terrestrial wildlife are briefly summarized below.

- Scientists in central Florida have been observing alligators from a contaminated lake and a control lake. The researchers have found that alligator eggs and newborns from the contaminated lake differ significantly from those in the control lake, showing reduced hatchability, reduced viability of offspring, endocrine "demasculation" of males, and "superfeminization" of females (Guillette et al. 1994). Juvenile alligators exhibit significantly smaller penis size (24 percent decrease) and lower plasma concentrations of testosterone (70 percent lower) when compared to alligators from the control lake (Guillette et al. 1996). The alligators from the contaminated lake have elevated levels of p,p'-DDE, a known endocrine disruptor, in their tissues, which researchers believe are associated with a large spill of a pesticide containing DDT. Studies into the mechanism of action of these effects are ongoing (Guillette et al. 1995, 1996).

- Researchers have been studying the mechanism of action of endocrine disruption in a common turtle species (the red-eared slider) and the African clawed frog (Palmer and Palmer 1995). Their work has focused on a potential biomarker for exposure to endocrine disruptors called vitellogenin (egg-yolk protein in the blood of egg-laying vertebrates). When stimulated by estrogen, the liver produces this protein and releases the protein into the bloodstream, where it then circulates to the ovaries and is deposited into an egg. Usually only females possess a sufficient amount of estrogen to produce vitellogenin; however, in a laboratory study, DDT induced vitellogenin production in male turtles and frogs. In another study, PCBs and lindane were found to induce estrogen receptor and vitellogenin accumulation in rainbow trout liver (Flouroit et al. 1995).

- In many egg-laying reptiles, the temperature of the incubating egg determines the sex of the offspring. PCBs applied to the shells of turtle eggs during the period of sexual differentiation counteracted male-producing temperatures and induced ovarian development (Bergeron et al. 1994). Further study of the mechanism of action and synergistic effects of different PCB congeners are ongoing (Crews et al. 1995).

- Researchers have recently been testing the hypothesis that endocrine disruptors, such as DDE, mercury, and PCBs, are playing a role in the decline of the endangered Florida panther population (Facemire et al. 1995). (Many have considered inbreeding the main factor up to this point.) A large percentage of males have exhibited abnormal reproductive organs, sterility, and production of abnormal or deformed sperm. Both males and females exhibit abnormal hormone ratios, with little difference in estradiol levels between males and females (i.e., evidence that males have been demasculated and feminized).

- The effluent from sewage treatment plants has been shown to induce vitellogenin synthesis is male fish (Folmar et al. 1996; Sumpter and Jobling 1995). The investigators attribute this effect to the estrogenic properties of environmental contaminants in the effluent, though specific chemicals were not identified.

In recent years, several organizations and governmental agencies, including EPA, have begun to support research efforts to further study endocrine disruptors. For example:

- The National Science and Technology Council, which advises the President and his Cabinet on directions for federal research and development efforts, has given EPA the task of developing a national research strategy on endocrine disruptors by 1998. EPA recently conducted two workshops to plan research in this area (see sidebar).

- The Chemical Industry Institute of Toxicology (CIIT) has developed a comprehensive research program to evaluate the potential for selected chemicals to affect the human endocrine system. One of the critical goals of this program is to understand the relationship between dose levels that produce an effect in laboratory cell cultures and the dose level needed for effects to be seen in laboratory animals.

- EPA's Risk Assessment Forum, which promotes scientific consensus on risk assessment issues, assembled a technical panel to study environmental endocrine disruptors. The panel has released a draft report that is intended to serve as an interim assessment and analysis of the environmental endocrine disruption hypothesis until a more extensive exploration of the issue can be completed by the National Academy of Science (U.S. EPA 1996d).

Research Planning Workshops on Endocrine Disruptors

***Purpose**:* To develop a national research strategy on endocrine disruptors, in response to growing public concern over their adverse effects.

***Findings**:* The hypothesis that endocrine disruptors cause a variety of adverse effects in wildlife and humans is of sufficient concern to warrant a concerted research effort. Research priorities include: identifying and characterizing effects on developing reproductive systems; and refining exposure assessments and research on toxicology of mixtures.

***Outcomes**:* EPA has published workshop findings and recommendations in scientific literature, begun implementing some of the identified research initiatives, and has formed an Endocrine Disruptor Research Coordination Committee.

Sources: Ankley et al. 1997; CENR 1996; Kavlok et al. 1996; U.S. EPA 1995b.

In addition to the recent focus on endocrine disruptors and their potential impact on ecological health, other research efforts relevant to potential ecological effects of the pollutants of concern include the following:

- EPA submitted a draft Mercury Study Report to the Science Advisory Board (SAB) for review in June 1996. The report was reviewed by SAB in February 1997. EPA expects to receive the opinion of SAB in the summer of 1997. The final Mercury Study Report will fulfill the requirements of CAA section 112(n)(1)(B), including a requirement to assess the environmental effects of mercury emissions.

- Dioxin has been classified as the most potent known animal carcinogen, and as a probable human carcinogen, since 1985. Increased concerns that dioxins in aquatic environments may be a major contributor to overall human dioxin exposure through fish and shellfish consumption, as well as increased evidence of its hazard to fish and wildlife, prompted EPA to reassess dioxin's effects on aquatic ecosystems. Work on characterizing ecological risks is in progress at EPA's Mid-Continent Ecology Division of the National Health and Ecological Effects Research Laboratory (NHEERL) in Duluth, Minnesota. EPA published the *Interim Report on Data and Methods for Assessment of 2,3,7,8-Tetrachlorodibenzo-p-dioxin Risks to Aquatic Life and Associated Wildlife* (U.S. EPA 1993a) on this research. The goal of the report is to review and evaluate relevant published and unpublished data and models currently available for analyzing dioxin exposure to and effects on aquatic life and wildlife. Information on related compounds, such as TCDF and PCBs, is not discussed in detail in the interim report; however, it is expected that the final report will assess the contribution of these related compounds to the risk for aquatic life and wildlife. The interim report findings are presented under the discussion of TCDD in this section.

Pollutants of Concern and Health Effects in St. Lawrence Estuary Beluga Whales

A 9-year epidemiological study in an isolated population of beluga whales documented higher levels of many pollutants of concern, including dieldrin, DDT, HCB, lead, mercury, PAHs, and PCBs, in these whales compared to Arctic belugas (Beland et al. 1993). The St. Lawrence whales exhibited high prevalence of tumors; high incidence of lesions to the digestive system, mammary glands, and other glandular structures; some evidence of immune system suppression; and frequent tooth loss and gum disease. No such lesions were observed in Arctic belugas or in other St. Lawrence aquatic mammals (e.g., seals). Researchers propose this case as a model for the potential long-term consequences of pollutants in the environment on human health (De Guise et al. 1995).

Based on information from the above research on ecological effects of pollutants of concern, as well as findings from recently published data, a summary of the potential ecological effects of the pollutants of concern is presented in Table II-8. For each effect attributable to a pollutant of concern, the table identifies the organism type(s) in which the effect has been observed (i.e., plants, invertebrates, fish, amphibians/reptiles, birds, and mammals). Table II-8 presents data on the following types of effects: death; cancer; reproductive/developmental, immunological, metabolic/enzyme, and neurological/ behavioral effects; damage to the kidney, liver, heart, lungs or gills, or gastrointestinal tract; exterior changes; and decreased growth or biomass. The information in this table is based on both field and laboratory studies. This report does not attempt to assess how the observed effects on individual animals contributes to changes in species populations within an ecosystem or how the laboratory studies relate to studies of effects observed in the environment. In addition, information on the exposure levels related to these effects is not provided.

Following the table, recent information on the ecological effects of the pollutants of concern is discussed by pollutant group. The ecological effects of nitrogen (i.e., eutrophication and its consequences) also are discussed in this section.

MERCURY AND COMPOUNDS

The data presented in Table II-8 summarize the potential effects of mercury exposure on plants, invertebrates, fish, birds, and mammals. A brief summary of some recent research findings is presented below:

♦ The effects of mercury on plants may include death, growth inhibition, leaf or root damage, chlorophyll decline, and reduced photosynthesis (Godbold 1991; Lindquist 1991; Schlegel et al. 1987).

♦ In fish and aquatic invertebrates, mercury exposure can cause death, reduced reproductive success, impaired growth and development, and behavioral abnormalities (U.S. EPA 1985).

TABLE II-8
Potential Effects of the Pollutants of Concern on Aquatic Life and Wildlife

Pollutant of Concern	Potential Ecological Effects[a]					
	Death	Cancer	Reproductive/ Developmental[b]	Immunological	Metabolic/ Enzyme	Neurological/ Behavioral
Cadmium and compounds	I F		I F B		F B M	I B
Chlordane	I F A B M	M	I F A B M	M	I M	F B M
DDT/DDE	P I		I B A M	M		F B
Dieldrin	B		A			B
HCB	P I F B		I F			I B
α-HCH						
Lead and compounds	I F A B M		I F A B M	F A	I F B M	F A B M
Lindane	I		I	F		I
Mercury and compounds	I F A B M	M	I F A B M	B	P F B	I F B M
PCBs	P I F B M	M	I F B M	F M	F B M	F B M
POM (PAHs)	I F M	F A M	I F B M	F B M	F	I F
TCDD (dioxins)	F B M		F B M	M		B
TCDF (furans)						
Toxaphene	I F A B M	M	I F B M			A F B

[a] Blank areas indicate that no data were found correlating the effect with the pollutant of concern (as opposed to data found indicating no correlation between the effect and the pollutant of concern).
[b] Includes endocrine-disrupting effects.

Key: P = Plants I = Invertebrates F = Fish A = Amphibians/Reptiles B = Birds M = Mammals

Sources: Arkoosh et al. 1994; Baturo et al. 1995; Constable and Orr 1994; Di Pinto et al. 1993; Doust et al. 1994; Dunier and Siwicki 1994; Eisler 1985, 1986a, 1986b, 1987a, 1987b, 1988, 1990; Eisler and Jacknow 1985; Ferrando et al. 1995; Fitchko 1986; Geyer et al. 1993; Government of Canada 1994; Hermsen et al. 1994; Hill and Nelson 1992; Hugget et al. 1992; Johnson et al. 1993; Lahvis 1995; Malbouisson et al. 1994; Schulz and Liess 1995; Tidou et al. 1992; Trust et al. 1994; and U.S. EPA 1993a, 1993b, 1993c.

TABLE II-8
Potential Effects of the Pollutants of Concern on Aquatic Life and Wildlife
(continued)

Pollutant of Concern	Potential Ecological Effects[a]						
	Kidney Damage	Liver Damage	Heart Damage	Lung/Gill Damage	Gastrointestinal Damage	External Damage	Decreased Growth/Biomass
Cadmium and compounds	B		B				I F B
Chlordane	M	M	M	M	M		I M
DDT/DDE							P
Dieldrin						B	
HCB		F B					P I B
α-HCH							
Lead and compounds[b]	F B	F B		F	A M	F B	I M
Lindane							I
Mercury and compounds[b]	B M	B		M			I F B
PCBs	F M	B M			B M	F B M	P I F B
POM (PAHs)[b]	M	F M			M		F B
TCDDs	M	F M		M	M	F M	F
TCDFs							
Toxaphene	F B				F	F B	I F B

[a] Blank areas indicate that no data were found correlating the effect with the pollutant of concern (as opposed to data found indicating no correlation between the effect and the pollutant of concern).
[b] Also causes other noncancer effects in plants, such as leaf and root damage, chlorophyll decline, and reduced photosynthesis.

Key: P = Plants I = Invertebrates F = Fish A = Amphibians/Reptiles B = Birds M = Mammals

Sources: Arkoosh et al. 1994; Baturo et al. 1995; Constable and Orr 1994; Di Pinto et al. 1993; Doust et al. 1994; Dunier and Siwicki 1994; Eisler 1985, 1986a, 1986b, 1987a, 1987b, 1988, 1990; Eisler and Jacknow 1985; Ferrando et al. 1995; Fitchko 1986; Geyer et al. 1993; Government of Canada 1994; Hermsen et al. 1994; Hill and Nelson 1992; Hugget et al. 1992; Johnson et al. 1993; Lahvis 1995; Malbouisson et al. 1994; Schulz and Liess 1995; Tidou et al. 1992; Trust et al. 1994; and U.S. EPA 1993a, 1993b, 1993c.

- Effects of mercury on birds may include death, liver and kidney damage, neurobehavioral effects, impaired growth and development, and reproductive effects (Eisler 1987a; MDNR 1993; Scheuhammer 1987, 1991). Reproductive effects are the primary concern for avian mercury poisoning and may occur at dietary concentrations well below those that cause overt toxicity.

- Extensive research on the toxicity of mercury to mammals indicates that effects vary depending on the form of mercury ingested, with methylmercury being the most toxic form. Methylmercury ingestion by mammals may cause death, neurological and behavioral effects, and damage to the heart, lung, liver, kidney, and stomach (ATSDR 1992e).

Population-level Effects of Mercury

Studies conducted on various communities have shown mercury to: (1) reduce species diversity of freshwater, brackish-water, and soil microbial communities; (2) reduce carbon fixation in phytoplankton communities; and (3) change the species composition of phytoplankton in an aquatic community. Although clear causal links between mercury contamination and population declines in various wildlife species have not been established, mercury may be a contributing factor to population declines of the endangered Florida panther and the common loon. Other researchers have concluded, however, that mercury levels in most areas are not sufficient to adversely affect bird populations.

Sources: Barr 1986; Ensor et al. 1992; FPIC 1989; Roelke et al. 1991.

OTHER METALS

Cadmium. As shown in Table II-8, adverse effects from exposure to cadmium have been demonstrated in invertebrates, fish, and birds. Recent studies have focused on the toxicity of cadmium to aquatic species (Kraak et al. 1992; Weinstein et al. 1992) and the identification of potential indicator species for cadmium exposure (Naimo et al. 1992). For example, researchers concluded that accumulation of some metals contributes to lesions to the shell of blue crabs in the Albemarle-Pamlico Estuarine System; however, the researchers did not observe this result for cadmium and lead (Weinstein et al. 1992).

Lead. Many noncancer ecological effects are associated with lead exposure, as shown in Table II-8. However, the availability of recent studies on the ecological effects of lead is limited. Rather, there is a strong interest currently focused on the neurological and behavioral effects in infants and children (see Section II.D). One recent study of fish demonstrated that exposure to waterborne lead may result in multiple effects on reproductive behavior and overall reproductive success in fathead minnows (Weber 1993). A recent review of wildlife contamination in Chesapeake Bay cited older articles that found that lead adversely affected embryo development and neurological behavior in the green and bull frogs (Heinz et al. 1992).

COMBUSTION EMISSIONS

As discussed in Chapter I, this pollutant group is comprised of 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD, dioxins), 2,3,7,8-tetrachlorodibenzofuran (TCDF, furans), and polycyclic organic matter (POM). Polychlorinated biphenyls (PCBs) are also discussed under this pollutant group; they were historically produced for use in electrical equipment and hydraulic fluids but are often now released into the environment in combustion gas emissions when PCB-containing materials are incompletely burned. The potential ecological effects associated with these pollutants are discussed below.

TCDD. As shown in Table II-8, dioxins are associated with effects in fish, birds, and mammals, including reproductive effects and other noncancer effects. EPA's assessment of ecological effects in aquatic life and associated wildlife discusses the effects of TCDD in detail (U.S. EPA 1993a). A brief summary of some of the findings is presented below; please refer to this document for the primary references for these studies.

- Available laboratory toxicity information indicates that aquatic invertebrates, plants, and amphibians may be substantially less sensitive to TCDD than fish. The report emphasized that the data are limited.
- The data indicate that young fish may be more sensitive to TCDD than older fish. Fish fry were most sensitive to effects from TCDD following exposure of eggs before or shortly after fertilization.
- In mammals and birds, the primary effect of concern is reproductive impairment. In addition, because of the bioaccumulation of TCDD in aquatic food webs, the species most exposed to TCDD are those species whose diet consists mostly of fish.

Ecological Effects of TCDD in Lake Trout Population in Lake Ontario

Commercial over-fishing and sea lamprey predation are thought to be the primary causes of lake trout decline in Lake Ontario. However, several findings suggest that TCDD toxicity could have contributed to the reproductive failure of stocked lake trout in Lake Ontario in the post-1950s: (1) the strong association between the occurrence of blue-sac disease (a stress syndrome) in sac-fry from TCDD-exposed fish in laboratory studies and from eggs collected from Lake Ontario; (2) the historical record of lake trout exposure to TCDD and related chemicals in Lake Ontario; and (3) the consistency of predicted toxicity with the field sampling record of no natural reproduction until 1986. Attainment of a self-sustaining population of lake trout in Lake Ontario requires exposures to TCDD and related chemicals below the level that causes significant reproductive impairment, in addition to sea lamprey control, maintenance of adequate spawning beds and water quality, and introduction of lake trout strains appropriate for Lake Ontario's conditions and food web.

Source: U.S. EPA 1993a.

One of the main issues of current interest is the mechanism of action of dioxins (i.e., how they bring about the effects). The unusual potency of dioxins in eliciting toxic effects suggested to researchers that a dioxin receptor existed. Based on a substantial amount of biological and genetic evidence, an intracellular protein called the aryl hydrocarbon receptor (Ah receptor) is believed to mediate biological responses to TCDD and related compounds. Figure II-2 presents a simplified diagram of how biological responses to dioxin-like compounds are mediated by the Ah receptor. Although our understanding of the Ah receptor is limited, mammals (including humans), birds, and fish all have exhibited detectable concentrations of the Ah receptor in a number of different tissues. In laboratory mammal studies, binding of dioxins to the Ah receptor has been associated with weight loss, edema, liver damage, promotion of tumors, and adverse effects on the immune and reproductive systems (Fox 1993). Researchers believe that responses to TCDD are likely to vary within and between species, as well as between tissues in individual species, based on differential responses to Ah-receptor binding. Several proteins are believed to contribute to TCDD's gene regulatory effects; the response to TCDD probably involves a complex relationship between multiple genetic and environmental factors.

Recent studies of dioxins and furans that were found through the literature search conducted for this Report to Congress demonstrated reproductive effects (e.g., embryo anomalies, decreased hatching success) in nesting wood ducks in an Arkansas bayou (White and Hoffman 1995) and potential neurotoxic effects in great blue heron hatchlings, as indicated by asymmetrical brain development (Henschel et al. 1995).

PCBs. As shown in Table II-8, PCBs are associated with adverse effects in plants, invertebrates, fish, birds, and mammals. These effects include reproductive/developmental, metabolic/enzyme, and neurological/behavioral effects. The specific effects and the levels at which these effects are seen may vary among the different PCB congeners and mixtures. Recent articles on the ecological effects of PCB exposure are summarized below. These studies focused on reproductive effects in invertebrates, fish, birds, and mammals, immune effects in fish and aquatic mammals, and other effects in plants and fish.

FIGURE II-2
Role of Ah Receptor in Biological Responses to Dioxin Exposure

Dioxin Exposure
↓
Free Dioxin in Tissues
↓
Dioxin Binding to the Ah Receptor in Cells
↓
Ah Receptor-Dioxin Complex Binding with DNA
↓
Gene Regulation
↓
m-RNA Regulation
↓
Protein Synthesis
↓
Biochemical Alterations
↓ Interactions of
↓ Multiple Target
↓ Genes
Early Cellular Responses (cell growth stimulation)
↓
Late (irreversible) Tissue Response (cancer, deformity)

Source: Adapted from EPA 1994c.

♦ *Reproductive and Developmental Effects*. A group of related studies investigated the reproductive effects of commercial PCB and combinations of PCB congeners in mink. Two of these studies found that commercial PCBs and combinations of PCB congeners adversely affected reproduction, but that exposure to single PCB congeners did not produce these same effects (Bäcklin and Bergman 1992; Kihlström et al. 1992). One of these studies showed a difference in adverse reproductive effects (e.g., late fetal death versus early fetal death) between two different commercial PCBs, Clophen A50 and Aroclor 1254 (Bäcklin and Bergman 1992). In another study, PCBs were found to reduce egg production and total reproductive capacity in a small crustacean species under laboratory conditions (DiPinto et al. 1993). A study that investigated the developmental toxicity of PCB congeners in minnows determined that several congeners caused severe teratogenic effects (e.g., inhibition of yolk absorption) and early hatching (Silberhorn et al. 1992). Recent studies of Great Lakes bird populations (e.g., common terns) provided further evidence of the developmental problems linked to PCBs during the 1980s (Becker et al. 1993; Hoffman et al. 1993; Yamashita et al. 1993). A literature review of wildlife contamination in Chesapeake Bay cited historical evidence of elevated post-hatch mortality in the leopard frog and in unspecified toad species related to PCB exposure (Heinz et al. 1992).

- *Immune System Effects*. In a recent study, production of antibodies was suppressed in juvenile chinook salmon exposed to the commercial PCB mixture, Aroclor 1254 (Arkoosh et al. 1994). In another study, delayed immune system responses in harbor seals fed herring from the Baltic Sea were associated with PCB exposure (Ross et al. 1995). In a study of dolphins, decreased immune system response was associated with elevated levels of DDT and PCBs in their blood (Lahvis et al. 1995).

- *Other Noncancer Effects*. Researchers have correlated detrimental effects on the quality and quantity of growth, as well as increased mortality, in aquatic plants with concentrations of PCBs (Doust et al. 1994). One study of winter flounder did not find a strong association between exposure to PCBs and the development of particular liver lesions (Johnson et al. 1993). Some recent studies have shown that effects from PCB exposure may be amplified in the presence of other chemicals. For example, a study in quail found that high levels of cadmium can amplify both the quantitative and qualitative retention of PCB congeners in muscle tissue, especially some of the congeners that are most toxic and resistant to metabolic degradation (Leonzio et al. 1992).

POM. The data in Table II-8 indicate that POM is associated with a range of adverse effects to aquatic and terrestrial wildlife. Studies on the adverse ecological effects from exposure to PAHs (a subset of POM) that have been completed in the last few years are summarized below. These studies examined immune effects in fish and birds as well as various other effects in algae, aquatic invertebrates, fish, and birds.

- *Immune System Effects*. In a recent study, production of antibodies was suppressed in juvenile chinook salmon exposed to PAHs (Arkoosh et al. 1994). Another study found that PAH exposure suppressed the immune system in both adult and young starlings, although only young starling showed overt signs of general toxicity (e.g., decreased body weight and blood hemoglobin concentration) (Trust et al. 1994). A study of effects on fish from PAH exposure found that macrophage activity, an important component of the cellular immune system (i.e., protects the host by eliminating foreign material), was markedly reduced (Huggett et al. 1992). The study results suggested that these effects may be reversible.

- *Other Noncancer Effects*. Recent research has investigated the increased toxicity of PAHs following exposure of the pollutants to solar radiation. One study that investigated the photo-induced toxicity of anthracene (a PAH compound) to a species of green algae showed that the combination of ultraviolet (UV-A) radiation and anthracene produced significant toxic effects. The study also concluded that algae may be slightly more resistant to photo-induced toxicity than fishes and invertebrates (Gala and Giesy 1992). Two recent studies on fish suggest a strong association between exposure to PAHs and the development of internal lesions, commonly found in the liver and kidney, as well as external lesions, such as lens cataracts (Huggett et al. 1992; Johnson et al. 1993). One of these studies found a strong correlation between sediment PAH concentration and stimulation of enzyme activity, which suggests that the PAHs are a main component of the adverse effects observed in the fish (Huggett et al. 1992). In another study, dramatic declines in PAH concentrations in sediment following the closure of a steel facility were followed by a decreased incidence of liver cancer and liver lesions in brown bullhead catfish (Baumann and Harshbarger 1995). A study of aquatic insect larvae from a contaminated area of the Niagara River found that higher body burdens of PAHs were associated with menta ("teeth") deformities. The researchers determined that the deformities were not passed on to future generations (i.e., that PAHs caused malformations but did not permanently alter genes or chromosomes) (Dickman et al. 1992).

PESTICIDES

Information on the adverse effects of the pesticides of concern in the Great Waters (chlordane, dieldrin, α-HCH, DDT/DDE, HCB, lindane, and toxaphene) is summarized in Table II-8. These data suggest that the pesticides of concern may produce a wide range of adverse effects in terrestrial and aquatic wildlife of the Great Waters. Recent literature on the ecological effects of pesticides was available mainly for DDT/DDE, HCB, and lindane; this information, except for information on the potential for endocrine disrupting effects (which was discussed earlier in this section), is summarized below.

DDT. Much of the recent literature on the ecological effects of DDT (and DDE) focuses on reproductive impairment in birds. Other studies demonstrate effects to the immune systems of mammals and impaired growth and survival of aquatic plants following DDT exposure.

♦ *Reproductive and Developmental Effects*. Recent studies on natural bird colonies in the Great Lakes indicate a decreased role of DDT/DDE in the reproductive success of these populations (Weseloh et al. 1994; Weseloh and Ewins 1994; Bowerman et al. 1995). One of these studies determined that for herring gull colonies in Lake Superior, eggshells were only eight percent thinner than before the introduction of DDT (Weseloh et al. 1994). In this study, reproductive failure was attributed to causes other than toxic contamination (e.g., predation and shortage of food supply). Similarly, another study attributed increasing populations of double-crested cormorants in Lake Ontario to reduced levels of contaminants (especially DDT/DDE) and increased availability of forage fish (Weseloh and Ewins 1994). A study investigating the combined effects of DDE and decreased food intake on reproduction in ringed turtle doves found that the lower the percentage of normal food intake, the greater the effects of DDE exposure on breeding success. DDE restricted breeding success apparently by limiting levels of hormones necessary to develop and maintain active gonads, adequate courtship and brooding behavior, and functional crop glands (Keith and Mitchell 1993). A study of freshwater pond snails, which are an important component of invertebrate fauna in most eutrophic and mesotrophic lakes, found that DDT exposure reduced their reproductive output (Woin and Brönmark 1992).

♦ *Immune System Effects*. In a study of dolphins, decreased immune system response was associated with elevated levels of DDT and PCBs in their blood (Lahvis et al. 1995).

♦ *Other Noncancer Effects*. In one study in aquatic plants, exposure to DDE caused mortality and had detrimental effects on the quality and quantity of growth (Doust et al. 1994).

Hexachlorobenzene. Recent studies on ecological effects from HCB exposure demonstrate reproductive and neurological effects in aquatic plants and animals. Impaired growth and survival of aquatic species and liver damage in fish and birds have also been shown.

- *Reproductive and Developmental Effects*. Reproductive effects from HCB exposure documented in recent literature include reduced reproduction in protozoa and fathead minnows and reduced fertility in waterflea crustaceans (Constable and Orr 1994; Government of Canada 1994).

- *Neurological/Behavioral Effects*. In freshwater snails, exposure to HCB inhibited body growth, altered metabolic activity, and stimulated egg production, which researchers attributed to neurotoxic effects (Baturo et al. 1995). Kestrels, a small falcon species, have shown ruffling of feathers and tremors following exposure to HCB (Government of Canada 1994).

- *Other Noncancer Effects*. In aquatic species, HCB was lethal to some marine invertebrates and significantly reduced the survival rates of freshwater caddisfly larvae and fathead minnows (Constable and Orr 1994; Government of Canada 1994; Schulz and Liess 1995). In another study, exposure to HCB caused mortality and had detrimental effects on the quality and quantity of growth in aquatic plants (Doust et al. 1994). One report documented the following other effects in aquatic species related to HCB exposure: reduced production of chlorophyll, dry matter, carbohydrate and nitrogen in some algae; decreased growth of algae and protozoa; digestive gland damage in crayfish; and liver necrosis in largemouth bass (Government of Canada 1994). In birds, HCB exposure induced porphyria (a group of disorders related to altered metabolism of breakdown products of hemoglobin known as porphyrins), increased liver weight, and slightly damaged livers in Japanese quail and produced significant weight loss, increased liver weight, and decreased heart rate in kestrels (Government of Canada 1994).

Lindane. Recent studies show effects in aquatic species following exposure to lindane, including reproductive effects in aquatic invertebrates, immune effects in fish, and behavioral effects in aquatic invertebrates.

- *Reproductive and Developmental Effects*. One study documented reduced reproduction in waterflea crustaceans from lindane exposure (Ferrando et al. 1995).

- *Immune System Effects*. Laboratory experiments on rainbow trout found that antibody production was significantly suppressed from lindane exposure at doses comparable to those found in fish in polluted fresh waters, and that as the dose increased the more the immune system was suppressed (Dunier and Siwicki 1994; Dunier et al. 1994).

- *Neurological/Behavioral Effects*. As a result of brief exposures to high concentrations of lindane, the mating behavior of freshwater crustaceans was disrupted (Malbouisson et al. 1994). In another study, the common mussel, *Mytilus edulis*, showed lower rates of feeding-type behavior after exposure to lindane-contaminated sediments (Hermsen et al. 1994). Also, an annelid worm exposed to lindane in seawater exhibited delays in settlement and metamorphosis; this species is otherwise known to be a colonizer of disturbed or polluted areas (Hill and Nelson 1992).

♦ *Other Noncancer Effects*. In two different studies, survival of a waterflea crustacean species was inhibited by lindane exposure (Ferrando et al. 1995; Tidou et al. 1992); in one of these studies, lindane also significantly repressed growth in the waterflea crustaceans (Ferrando et al. 1995). In studies comparing data for several species of fish, acute toxicity of lindane decreased with increasing total body fat in fish (Geyer et al. 1993, 1994).

NITROGEN COMPOUNDS

The availability of biologically-usable nitrogen normally limits biological productivity in coastal waters, but an over-abundance of nitrogen is a concern in areas where nutrient enrichment problems, known as eutrophication, have developed. In addition to increasing productivity, nutrient enrichment generally alters the normal ratios of nitrogen to phosphorus and to other elements, such as silicon. This alteration may induce changes to phytoplankton community structure. Species that normally occur in low abundance may be favored, and in some cases, toxic and/or noxious algal blooms may result. On the New England coast, for example, the number of red and brown tides and shellfish problems from nuisance and toxic plankton blooms have increased over the past two decades. In coastal areas with poor or stratified circulation patterns (e.g., Chesapeake Bay, Long Island Sound), the "overproduced" algae tends to sink to the bottom and decay, using all (anoxia) or most (hypoxia) of the available oxygen in the process, killing or driving away organisms that require oxygen. In addition, the increase in suspended matter due to overproduction of algae decreases light penetration, causing a loss of underwater seagrass and coral communities.

Atmospheric deposition of nitrogen compounds is recognized by all U.S. estuarine programs on the east coast as either a significant contributor to estuarine eutrophication or a mechanism of possible concern (ECARA 1996). An important consideration for controlling atmospheric deposition of nitrogen is that the region from which the atmospheric nitrogen pollution arises (i.e., the "airshed") is larger than the watershed that drains into the waterbody and much larger than the water surface that is potentially affected. The extent of "airsheds" is now starting to be recognized (e.g., see the discussion of the Chesapeake Bay airshed in Section C of Chapter IV).

II.D Human Health Effects

This section is intended to provide a brief overview of the current literature on the potential human health effects from exposure to the Great Waters pollutants of concern. In general, the information presented in this section represents data published since the background document from the First Report to Congress on exposure and effects (Swain et al. 1992a) was completed through 1995. Because the information presented in this report covers only recent studies, it cannot be used alone to determine whether these effects are widespread in the environment. As in the First Report to Congress, the contribution of atmospheric deposition to toxic contamination and potential human health effects associated with exposure to the pollutants cannot be quantified at this time.

Much of the information presented in this section on human health effects is based on effects observed in animals, mainly in laboratory animal studies, that are suggestive of potential human health effects. Unlike the effects of certain pollutants of concern on ecological health in the Great Waters (discussed in Section II.C), data on adverse health effects observed in humans from non-occupational exposure to the pollutants of concern are limited. This section focuses on the effects of the pollutants of concern and does not attempt to summarize the exposure levels responsible for the observed effects. In addition, this section does not attempt to distinguish effects that may occur with long-term exposure to the pollutants of concern from those effects caused by acute, high-level exposures (e.g., accidental spills). The potential effects of a pollutant may vary with duration of exposure, possibly due to a breakdown of the chemical in the body to another chemical that is more toxic or affects other target organs than the exposed chemical.

Conclusions from the First Report to Congress

Information from the First Report to Congress is presented here to provide a foundation for the subsequent discussion of the recent information available for this report. The research findings and studies presented in the First Report to Congress, as well as the background document on exposure and effects (Swain et al. 1992a), led to the following conclusions regarding the potential human health effects from exposure to the pollutants of concern:

♦ Numerous studies indicated potential human health effects associated with the pollutants of concern (see sidebar).

♦ Though many of the pollutants of concern are probable carcinogens, the noncancer effects of these pollutants are also a significant concern and may be as detrimental as cancer, or more so, to individuals and populations.

♦ It is possible for low-level exposure to several of the pollutants to have little or no measurable effect on an adult, yet alter the formation and function of critical physiological systems and organs in

Potential Human Health Effects Caused by Pollutants of Concern

- Cancer
- Reproductive effects
- Developmental effects, including effects on embryos, fetuses, and children
- Neurological (i.e., brain and nervous system) effects
- Immune system effects
- Endocrine system effects, including effects on hormone synthesis and function
- Other noncancer effects, including liver and kidney damage

children of the exposed adult, especially when the child is exposed at critical developmental stages in the mother's womb.

- ♦ Some human health effects caused by the pollutants of concern are subtle, result from long-term exposures to low levels of pollutants, and may be delayed in onset and occur across several generations. For example, long-term exposure to low levels of mercury may result in kidney or nervous system damage only after gradual exposure and bioaccumulation in the body.

The remainder of this section presents updated information on human health effects associated with the Great Waters pollutants of concern.

Current Understanding of Human Health Effects

Since the First Report to Congress, updated information on the potential human health effects from exposure to the pollutants of concern has become available. This section summarizes the information first by discussing some notable research efforts relevant to the potential human health effects of some Great Waters pollutants of concern, followed by a brief overview of recent data on the potential human health effects for each pollutant of concern.

As discussed in Section II.C, the role of endocrine disruptors in causing adverse effects in wildlife and humans is an emerging and controversial issue. Effects of endocrine disruptors have been seen mostly in wildlife and laboratory experiments; however, there are a few known cases of accidental exposure of humans to high concentrations of endocrine disruptors (see sidebar for an example). Humans are especially susceptible to adverse effects when exposure to endocrine disruptors occurs during periods that are tightly controlled by hormonal activity, such as during embryo and fetal development (Bern 1992). As an example, the chemical hormone diethylstilbestrol (DES) was given to millions of women to prevent miscarriages between the 1940s and 1970s. Though the women were largely unaffected by their exposure to DES, their children exhibited adverse reproductive effects such as decreased fertility in both sexes, testicular cancer in males, and abnormal pregnancies in females (Colborn et al. 1993; Hileman 1994). Scientists use the DES incident as a model of how endocrine disruptors may affect humans.

> **Human Exposure to Endocrine Disruptors**
>
> Accidental exposure to cooking oil contaminated with PCBs occurred in Japan in 1968 and Taiwan in 1979, affecting 3,000 to 5,000 people overall. Children born to women who consumed the oil when pregnant exhibited birth defects and developmental and behavioral deficiencies that scientists believe may be related to hormonal changes caused by exposure (Lai et al. 1993; Rogan and Gladen 1992). Upon reexamination of the Taiwanese children in the early 1990s, researchers found that the children scored lower than controls on IQ tests, girls were shorter than average, and boys' penises were significantly smaller than normal (Chen and Hsu 1994; Guo et al. 1993).

Some researchers have cautioned that because of the complex interactions involved, proving a cause and effect link between adverse effects in humans and endocrine disruptors will be difficult. Some raise doubts about the severity of the effects of endocrine disruptors shown in some of the scientific literature (Safe 1995). The investigators note that these chemicals are only weak hormones; natural estrogens have been shown to be over a thousand times stronger than estrogenic pesticides (Birnbaum 1994). In addition, some researchers theorize that the net effect of natural and chemical endocrine disruptors may be zero, especially given that some environmental estrogens are found in plants and have been shown to have beneficial effects, such as inhibiting tumor formation (Safe 1995). The significance of endocrine-disrupting effects in humans remains unclear.

Recent work on mixtures of endocrine disruptors raises possible answers to some of these arguments. Researchers found that, in a simple yeast estrogen system, combinations of two weak endocrine disruptors, such as dieldrin and toxaphene, were 10 to 1,600 times more potent than individual compounds in eliciting binding and activation of the human estrogen receptor (Arnold et al. 1996). This research is preliminary, but has important implications, especially because pollutants in the environment are typically found in mixtures.

In addition to the recent focus on endocrine disruptors, other research efforts relevant to the potential human health effects of the pollutants of concern include the following:

- EPA submitted a draft Mercury Study Report to the Science Advisory Board (SAB) for review in June 1996. The report was reviewed by SAB in February 1997. EPA expects to receive the opinion of SAB in the summer of 1997. The final Mercury Study Report will fulfill the requirements of CAA section 112(n)(1)(B), including a requirement to assess the public health impacts of mercury emissions.

- In response to growing scientific controversy regarding the potential effects from exposure to dioxins and related compounds, EPA began a scientific reassessment of the health risks from exposure to TCDD and chemically-related compounds (known collectively as dioxins and including TCDFs) in April 1991. Activities under the dioxin reassessment included updating and revising the health assessment and exposure assessment documents and performing research to characterize ecological risks in aquatic ecosystems. The dioxin reassessment document was published in draft form in June 1994 and is found in two reports (each 3 volumes): the health assessment document (U.S. EPA 1994c) and the exposure document (U.S. EPA 1994d). More detailed information on the human health effects related to TCDDs and TCDFs are discussed later in this section.

Based on the above research areas of interest plus human health effects data from the First Report to Congress and from recently published studies, Table II-9 presents the potential human health effects associated with the Great Waters pollutants of concern, except nitrogen. Nitrogen compounds are not included in this table because nitrogen compounds that are atmospherically deposited generally are not a direct hazard to human health. (Nitrates in drinking water from wells are of concern in many areas, but are not linked to atmospheric deposition.) For balance in understanding the importance of nitrogen oxides and other nitrogen emissions to the atmosphere, effects on human health due to ozone formation resulting from nitrogen compounds are briefly discussed later in this section. The data in Table II-9 generally are based on a compilation of results from laboratory studies on whole animals, toxicity/cellular studies in "test tubes" (i.e., in vitro studies), and human epidemiological studies describing occupational or accidental exposure to high concentrations of chemicals. Although these studies identify serious potential effects for humans, it is difficult to determine the adverse effects that would actually occur with chronic and low-level exposure to these pollutants in the environment. Since the First Report to Congress, Table II-9 has been updated to recognize the

TABLE II-9
Potential Human Health Effects Associated With Pollutants of Concern [a]

Pollutant	Potential Effects on Human Health [b]					
	Cancer [c]	Reproductive/ Developmental	Neurological/ Behavioral	Immunological	Endocrine	Other Noncancer [d]
Cadmium and compounds	Probable	●	●	●		Respiratory and kidney toxicity
Chlordane	Probable	●	●	●	●	Liver toxicity
DDT/DDE	Probable	●	●	●	●	Liver toxicity
Dieldrin	Probable	●	●	●	●	Liver toxicity
Hexachloro-benzene	Probable	●	●	●	●	Liver toxicity
α-HCH	Probable					Kidney and liver toxicity
Lead and compounds	Probable	●	●	●	●	Kidney toxicity
Lindane	Probable [f]	●	●	●	●	Kidney and liver toxicity
Mercury and compounds	Possible [g]	●	●	●	●	Kidney toxicity
PCBs	Probable	●	●	●	●	Liver toxicity
Polycyclic organic matter [e]	Probable	●		●	●	Blood cell toxicity
TCDF (furans)	Not classifiable	●		●	●	Liver toxicity
TCDD (dioxins)	Probable [f]	●	●	●	●	Chloracne
Toxaphene	Probable	●	●	●	●	Cardiovascular effects; liver toxicity

[a] Sources: Barnea and Shurtz-Swirski 1992; Cassidy et al. 1994; Chowdhury et al. 1993; Clayton and Clayton 1994; Colborn et al. 1993; Howard 1991; Linder et al. 1992; Soto et al. 1993; ATSDR Toxicological Profiles (see list in References chapter); and U.S. EPA 1987a, 1987b, 1988, 1989, 1990, 1991a, 1993d, and 1994c.
[b] For purposes of this table, a pollutant was considered to induce an effect if human or laboratory mammal data indicating a positive result were available. Blanks mean that no data indicating a positive result were found in the references cited (not necessarily that the chemical does not cause the effect).
[c] Cancer classifications: (1) "probable human carcinogen" when there is limited or no evidence of human carcinogenicity from epidemiological studies but sufficient evidence of carcinogenicity in animals (corresponds to EPA weight-of-evidence category B); (2) "possible human carcinogen" when there is limited evidence of carcinogenicity in animals and inadequate or lack of human data (corresponds to EPA weight-of-evidence category C); and (3) "not classifiable as to human carcinogenicity" when there is inadequate human and animal evidence of carcinogenicity or when no data are available (corresponds to EPA weight-of-evidence category D). Data on cancer classifications are obtained from EPA's Integrated Risk Information System (IRIS), unless otherwise noted.
[d] This column reports only a sample of other noncancer effects that may occur as a result of chronic exposure to the pollutant. Additional adverse human health effects may be associated with each chemical.
[e] POM represents a class of numerous compounds; not every compound is responsible for the potential effects on human health.
[f] Data from Health Effects Assessment Summary Tables (HEAST), which classify these chemicals as probable human carcinogens; however, these carcinogenic evaluations are currently under review by EPA.
[g] Inorganic (mercuric chloride) and organic (methylmercury) forms are classified as "possible," whereas elemental is "not classifiable."

potential endocrine-disrupting effects of DDT/DDE and lindane, and the "possible human carcinogen" classification for mercury (organic and inorganic forms).

The current information on the human health effects associated with each of the pollutant groups is highlighted in the remainder of the section.

MERCURY AND COMPOUNDS

Mercury, a metal, is discussed separately from other metals because of the significant emphasis placed on mercury in section 112 of the CAA and the difference in its behavior and effects compared to other metals. It has long been known that organic mercury (methylmercury) bioaccumulates in fish, and can biomagnify in the food web. In the air, mercury exists primarily as elemental (Hg^0) and inorganic (Hg[II]) mercury. Most of the mercury in water, soil, or sediments occur in the form of elemental mercury and inorganic mercury salts.

Humans are most likely exposed to mercury indirectly as methylmercury through a diet containing contaminated fish, instead of directly from inhalation of mercury in air. Ingestion of mercury-contaminated fish can result in various health effects, particularly toxicity to the nervous system in adults and in children exposed as fetuses. As shown in Table II-9, mercury may also affect the reproductive and immune systems. Since the First Report to Congress, EPA has recommended the current rating of the scientific weight-of-evidence regarding the human carcinogenicity of mercury (U.S. EPA 1993d):

- Elemental mercury: not classifiable as to human carcinogenicity;
- Inorganic (mercuric chloride) and organic (methylmercury) mercury: possible human carcinogens.

In recent years, researchers, including EPA, have focused on several issues related to the health effects of methylmercury: (1) improving the quantitative estimate of the relationship between mercury levels in air and exposure levels; (2) using refined statistical approaches and the application of physiologically-based pharmacokinetic models to evaluate the critical dose levels at which health effects occur from mercury; and (3) effects on nervous system development in populations that consume mercury-contaminated fish. New data are available from a recently-published study investigating neurological effects in children belonging to a fish-consuming population in the Seychelles Islands. Data from evaluation of these children (up to six years of age) have been collected and are being analyzed. Data from a similar study in the Faroe Islands have been published in abstract form; however, further investigation is being conducted to determine if the study subjects also may have been exposed to PCBs. Smaller scale studies evaluating effects in populations around the Great Lakes also are in progress. The above data and methodologies have not yet been incorporated into an EPA risk assessment of methylmercury because a majority of the new data are either not yet published or have not yet been subjected to rigorous review.

OTHER METALS

Cadmium. As indicated in Table II-9, cadmium has been linked to numerous adverse human health effects, including respiratory and kidney toxicity, probable carcinogenicity, reproductive and developmental effects, and immunological effects. Recent literature discussing the adverse human health effects of exposure to cadmium is limited and generally is focused on how cadmium alters the function of the kidney, which is known to be the critical organ for cadmium exposures (WHO 1992). Cadmium exposure has been found to cause deficient vitamin D metabolism in the kidney, which subsequently affects the calcium balance and bone density and may result in osteoporosis or osteomalacia (both bone diseases characterized by a change in the mineral and matrix phases of bone tissue). Exposure to cadmium has also been linked to developmental effects (e.g., particularly low birth weight).

Lead. Recent literature on human health effects from lead exposure generally supports the findings presented in II-9: lead is considered a probable carcinogen and may affect reproduction and development, alter the immune, nervous, and endocrine systems, and damage the kidney. While the correlation between high levels of lead in blood and adverse human health effects is relatively well known, current research has focused on the adverse effects associated with low levels of lead in blood (<30 micrograms of lead per deciliter of blood, or μg/dL) (see sidebar). Low levels of lead may be found in blood of a significant portion of the general public (U.S. EPA 1991b). Effects from low level exposure range from subtle cellular changes, such as effects on red blood cell metabolism, to pronounced effects on physical and mental development (Hovinga et al. 1993; Huseman et al. 1992; Kim et al. 1995). Blood lead levels as low as 10 μg/dL (and possibly lower) may result in adverse human health effects (Mushak et al. 1989; U.S. EPA 1991b). EPA has also noted that infants and young children may be most susceptible to adverse effects resulting from lead exposures (U.S. EPA 1991b).

Low-Level Lead Exposure

Because of the adverse human health effects associated with low-level lead exposure coupled with the numerous exposure pathways for lead, EPA has abandoned its standard approach for evaluating chemical toxicity in favor of a site-specific modeling approach. In 1994, EPA released a revised version of the *Integrated Exposure Uptake Biokinetic Model for Lead in Children* as its recommended methodology for assessing lead exposure. The model considers the principal lead exposure pathways (e.g., drinking water, diet, inhalation) to estimate lead levels in blood of children. Overall lead intake is then integrated in the model to estimate blood lead concentration. This concentration can then be used to predict: (1) the percentage of the exposed population that will have blood lead levels greater than 10 µg/dL; or (2) the probability that a child exposed to this level will have a blood lead level greater than 10 µg/dL. EPA defines 10 µg/dL blood lead as the lower bound of the range that is known to cause adverse effects on behavior in young children.

Source: U.S. EPA 1994b.

COMBUSTION EMISSIONS

TCDD and TCDF. As shown in Table II-9, the potential human health effects from exposure to TCDD include probable carcinogenicity, and reproductive and developmental, neurological and behavioral, immunological, and endocrine system effects. For TCDF, the potential human health effects are reproductive and developmental, immunological, and endocrine system effects, and liver toxicity. The dioxin reassessment document (U.S. EPA 1994c) discusses the effects of TCDD and related (compounds collectively referred to as dioxins) in detail. A summary of some of the research presented in the reassessment is discussed below; please refer to the reassessment document for the primary literature references for these studies.

- *Carcinogenicity*. A number of new studies provide further evidence that dioxins are probable human carcinogens. Because available human studies alone cannot confirm whether there is a causal relationship between dioxin exposure and increased cancer incidence, assessors have extrapolated from available animal data to potential human cancer. Recent animal studies have demonstrated dioxins to be carcinogenic in hamsters and small fish. All of these data have contributed to the weight-of-evidence that dioxins and related compounds may be carcinogenic, under certain circumstances, in humans.

- *Reproductive and Developmental Effects*. The potential for dioxins and related compounds to cause adverse reproductive and developmental effects in animals has been recognized for many years. Recent laboratory studies have suggested that altered development may be among the most sensitive TCDD endpoint in laboratory animals, although the likelihood and level of response in humans is much less clear. One study of monkeys found that chronic exposure to TCDD increased the risk of endometriosis, a female reproductive system disorder that can be painful and produce infertility. Human epidemiologic studies on the relationship between dioxin exposure and endometriosis are planned as part of follow-up research to a 1976 dioxin release in Seveso, Italy.

- *Immune System Effects*. Recent animal studies confirm that dioxins may cause immunological effects and suggest that some effects may occur after low-level exposure. There is conflicting evidence, however, concerning the effects of these compounds on humans. A developing human immune system is believed to be particularly sensitive to the effects of exposure to dioxins compared to adults, but additional research is necessary to confirm this hypothesis.

- *Endocrine System Effects*. Two human epidemiologic studies linked exposure to TCDD with changes in male reproductive hormone levels. Two of three studies found decreased testosterone levels and one of two studies observed an increase in a female ovulation hormone in males. Animal studies have produced similar results.

- *Other Noncancer Effects*. Chloracne is a severe acne-like disorder that develops within humans after a few months of exposure to dioxin. It may disappear in some individuals after exposure is discontinued, or persist in others for many years. Limited data exist to determine the doses at which chloracne is likely to occur, but long lasting, high-intensity exposures that begin at an early age are believed to increase an individual's chances of contracting this skin disorder. Another potential effect is enzyme induction, which has occurred in animals exposed to TCDD compounds. The biochemical alteration may either benefit the animal or result in adverse effects to the animal (i.e., alter metabolism of certain chemicals by increasing or decreasing toxicity). Studies linking this effect directly to humans are not available.

Most dioxins are thought to affect animals and humans by binding with the Ah receptor. The Ah receptor, which has been detected in humans, and its mechanism of action are discussed in Section II.C.

PCBs. This section summarizes the potential human health effects that are generally common to all PCB compounds, although the levels at which these effects may occur vary between compounds. In addition, specific compounds may produce their own range of effects.

As shown in Table II-9, PCBs are classified as probable carcinogens, and potentially affect reproductive, neurological, immunological, and endocrine processes in the human body. Recent research suggests that PCBs may be able to act as endocrine disruptors in ways similar to many pesticides and dioxins (McKinney and Waller 1994).

PCBs remain a significant concern because, although they have not been produced in this country for over a decade, they are often released into the environment in combustion gas emissions when PCB-containing materials are incompletely burned. In addition, appreciable concentrations are still found in water, and animal and human tissue, milk, and blood. As reported in Section II.A, the majority of fish advisories in the Great Waters are due to the presence of PCBs.

One recent study comparing serum PCB levels in people who ate Great Lakes fish (i.e., consumers) to a control population found that serum PCB levels did not change substantially over a seven-year period (Hovinga et al. 1992), as shown in Table II-10. The study proposes a number of reasons for the static PCB levels in the fish-consuming population: (1) restrictions on PCB production alone may not ensure decreasing levels of PCB exposure in human populations; (2) other sources of PCB contamination may be major sources of exposure; or (3) seven years may not be a long enough time to see a decrease in body burdens of PCBs, due to the persistent nature of these compounds, in human populations.

Changes in Serum PCB Levels in Great Lakes Populations

Hovinga et al. (1992) compared data from a 1982 study that examined 115 individuals who consumed Great Lakes fish and 95 who did not eat fish (i.e., controls) to a similar study in 1989 that reevaluated these individuals. Comparison of the data from the two studies (Table II-10) shows a significant decrease in mean serum DDT levels in both fish-eaters (almost 40 percent) and controls (approximately 30 percent). In contrast, serum PCB levels decreased only slightly in fish-eaters and remained relatively constant in controls. These results support other studies that have found stable PCB levels over time when DDT levels have been decreasing and suggest that, despite their ban, restrictions on PCBs have not been as effective in reducing levels of contamination in humans as those placed on DDT.

TABLE II-10
Mean Serum PCB and DDT Levels in Fish Eaters and Controls (1982 vs. 1989) [a]

	Fish Eaters		Controls	
	1982	**1989**	**1982**	**1989**
Serum PCB (ppb)	20.5	19.0	6.6	6.8
Serum DDT (ppb)	25.8	15.6	9.6	6.8

[a] Values represent 111 fish eaters and 90 controls (out of original 115 and 95, respectively).

Potential sources of exposure to PCBs other than fish consumption are disposal of previously manufactured products containing PCBs; atmospheric transport from other countries where PCBs are still produced and used; and cycling of PCBs in the environment. The widespread use of PCBs in electrical equipment is of concern, but there has been an increased awareness of the combustion of waste containing PCBs. The continued detection of PCB concentrations in the environment suggests that there is a reservoir of PCBs from "old" pollution that cycles between sediments, water, and air.

In view of this evidence of continuing persistence of PCBs in the environment and in the human population, effects on humans related to this group of chemicals remain a concern and many studies assessing their effects are currently underway. A 1992 workshop sponsored by EPA documented several human health effects from exposure to PCBs (see sidebar). Other recent findings include:

♦ *Carcinogenicity*. In response to evidence that the development of cancer may involve miscommunication between cells, a recent study investigated the effect of PCB exposure on intercellular communication in human breast cells and found that cell communication was inhibited with increasing doses (Kang et al. 1996).

♦ *Reproductive and Developmental Effects*. In female rat pups given doses of PCBs, one study observed delayed puberty, fertility impairment, and irregular estrus cycle patterns (Sager and Girard 1994). The investigators suggested that these reproductive effects may be related to impairment of endocrine function. One study found that consumption of PCB-contaminated sport fish from Lake Ontario did not increase the risk for spontaneous fetal death in humans, which has been observed in various mammalian species after PCB exposure (Mendola et al. 1995).

♦ *Neurological/Behavioral Effects*. The effects of prenatal exposure to PCBs on neurological function in children were investigated in a follow-up study of children from the contaminated rice oil incident in Taiwan (introduced in the discussion of endocrine disruptors earlier in this section). Researchers concluded that exposure of the fetus to PCBs may impair the psychological functions of the brain in the child and that PCBs may persist in the brain for a long period (Chen and Hsu 1994).

Workshop on Developmental Neurotoxic Effects Associated With PCB Exposure

In September 1992, EPA sponsored a workshop on the developmental nervous system effects of PCBs. At this meeting, experts addressed whether currently available health effects data are sufficient to support developmental neurotoxicity data in risk assessment. Some current findings regarding human health effects associated with PCBs were discussed and are summarized below.

- Women exposed to PCBs through contaminated cooking oil experienced irregularities in menstrual cycles, which may suggest alteration in ovarian function.

- Potential nondevelopmental nervous system effects include headaches, numbness, altered peripheral nerve function, and decreased neurobehavioral function measured through visual memory and problem solving ability. In animals, PCB exposure has been shown to affect the actions of dopamine (a brain neurotransmitter).

- PCB exposure has been associated with atrophy of the thymus and immunosuppression in animals (i.e., inhibition of immune cells required for tumor resistance).

- Chloracne and liver dysfunction are associated with occupational exposure to PCBs in humans. In animal studies, increased mortality, skin ailments, hepatotoxicity, and weight loss have been demonstrated.

Source: U.S. EPA 1993b.

- *Other Noncancer Effects*. Prenatal exposure to PCBs was associated with lowered intellectual function in school-age children (Jacobsen and Jacobsen 1996). The most highly exposed children were three times as likely to have low average IQ scores and twice as likely to be at least two years behind in reading comprehension. Effects were associated only with prenatal exposure, even though larger quantities of PCBs are transferred by breast-feeding than across the placenta.

As discussed in the recently published health effects reassessment for dioxin, some PCBs have been found to produce similar effects as dioxins.

POM. As shown in Table II-9, adverse human health effects associated with POM include reproductive and immunological effects, as well as probable carcinogenicity. Health effects data on POM are available primarily for polycyclic aromatic hydrocarbons (PAHs), one group of POM chemicals. Recent literature on health effects of PAHs corroborates previous findings and investigates the mechanism of action for PAH effects.

- *Carcinogenicity*. There is some speculation that ultraviolet light acts as a cofactor in the development of PAH-associated skin cancer, although this hypothesis requires further evaluation (Saboori and Newcombe 1992).

- *Reproductive and Developmental Effects*. It has been demonstrated in human placenta samples that short-term exposure to two PAHs (benzo[a]pyrene and 3-methylcholanthrene) increases the secretion of human chorionic gonadotrophin (HCG) (an important hormone in the human placenta) in the first trimester. This effect was maintained after application of the PAHs was discontinued. The secretion of this hormone may be involved in adverse reproductive effects observed with these pollutants (Barnea and Shurtz-Swirski 1992).

- *Immune System Effects*. Exposure of experimental animals in the womb to PAHs was found to alter development of the immune system and cause severe and sustained postnatal immunosuppression (i.e., inhibition of immune cells necessary for tumor resistance) (Holladay and Luster 1994).

PESTICIDES

The pesticides of concern for the Great Waters are chlordane, DDT/DDE, dieldrin, hexachlorobenzene, α-HCH, lindane, and toxaphene. As shown in Table II-9, these pesticides are probable human carcinogens and potentially result in toxic effects to reproductive, immune, and endocrine systems, as well as other noncancer effects.

Much of the recent literature concerning adverse health effects from exposure to the pesticides of concern in the Great Waters discusses their estrogenic and other hormonal effects (see earlier discussion of endocrine disruptors), as well as mechanisms of action of these effects (Chowdhury et al. 1993; Foster et al. 1992a, 1992b; Johnson et al. 1992; Juberg and Loch-Caruso 1992; McNutt and Harris 1993). Current research that has further explored the mechanisms of action of the pesticides in producing their known effects, other than endocrine disruption, are summarized below.

- *Carcinogenicity*. In response to evidence that the development of cancer may involve miscommunication between cells, a few recent studies have investigated the effects and mechanism of action of cancer-promoting chemicals, including dieldrin, DDT, lindane, and toxaphene, on intercellular communication (Kang et al. 1996; Leibold and Schwarz 1993; Rivedal et al. 1994; Tateno et al. 1994). They have found that cell communication is inhibited with increasing doses of these chemicals.

♦ *Reproductive and Developmental Effects*. In addition to studies of reproductive effects associated with endocrine disruption, recent studies discuss the effects and accumulation of the pesticides in reproductive organs (e.g., ovary) (Bourque et al. 1994; Lindenau et al. 1994; Singh et al. 1992). One recent study concluded that DDT is not toxic to sperm (Linder et al. 1992).

♦ *Neurological/Behavioral Effects*. Studies on the adverse effects from exposure to neurotoxic pesticides provided further support of their adverse neurological effects, investigated the mechanisms of action producing these effects, the specific regions of the central nervous system affected (e.g., the motor primary cortex and hippocampus in the brain), and whether there is a critical exposure period for these effects (Barrón et al. 1993; Eriksson et al. 1992, 1993; Gilbert and Mack 1995; Goldey and Taylor 1992; Gopal et al. 1992; Johannson et al. 1993; Kilburn and Thorton 1995; Nagata et al. 1994; Rivera et al. 1992). Many of these studies evaluated nervous system effects during development in young animals, and determined that progression of the effects, as well as severity of these effects, is influenced by the time of exposure (e.g., formation of the nervous system is a sensitive stage of development). Data also suggest that some effects of early exposure to pesticides on certain regions of the central nervous system may be reversible.

♦ *Immune System Effects*. Studies investigating the mechanisms of action for producing immunological effects and the effects on specific organs in the immune system (e.g., spleen) have confirmed the potential adverse immunological effects related to these pesticides. Immune system effects (e.g., suppression, autoimmunity) were observed in humans with elevated levels of chlordane (McConnachie and Zahalsky 1992). Animal data have demonstrated that immune system stimulation (i.e., increased antibodies) and/or suppression (e.g., decreased production of certain antibody-forming cells) may result with exposure to DDT, dieldrin, HCB, and lindane (Flipo et al. 1992; Meera et al. 1992; Rehana and Rao 1992; Saboori and Newcombe 1992; Saha and Banerjee 1993; Schielen et al. 1993).

♦ *Other Noncancer Effects*. A review of literature on human health effects from long-term exposure to pesticides concluded that (1) chlordane may cause disorders in bone marrow; (2) DDT can cause chloracne, chromosome aberrations, tremors, muscular weakness, and high levels of cholesterol and triglyceride; and (3) hexachlorobenzene may cause metabolic abnormalities and liver cancer (Maroni and Fait 1993). Another study demonstrated that lindane may impair development of blood cells in the bone marrow (Parent-Massin and Thouvenot 1993).

NITROGEN COMPOUNDS

Nitrogen in the molecular form N_2 is the most abundant gas in the earth's atmosphere, and is essentially chemically inert under ambient environmental conditions. Other compounds of nitrogen, particularly oxides of nitrogen, are common in the air due to their formation mainly through coal and oil fossil-fueled electric power generation and automobile fossil fuel combustion, and produce a variety of effects on human health and the environment. EPA has several programs, in addition to the Great Waters program, that are evaluating and attempting to reduce the threat to human health from atmospheric nitrogen dioxide, ozone, and acidic precipitation or particulates (U.S. EPA 1995c, 1995d, and 1995e). These other programs also examine general environmental effects of the nitrogen compounds, while the Great Waters program concentrates on the effects from nutrient enrichment, or eutrophication, in waterbodies from atmospheric deposition of nitrogen compounds (see Section II.C on ecological effects).

Oxides of nitrogen are produced abundantly by many modern combustion processes. Of the oxides of nitrogen, nitrogen dioxide (NO_2) is the most abundant in emission plumes or vehicle exhaust. At times, NO_2 itself can reach ambient concentrations associated with a variety of acute and chronic health effects. A more common problem is that, in the presence of sunlight, nitrogen oxides in the atmosphere react with volatile organic compounds (VOCs) to form ozone. Although ozone in the stratosphere is essential for protecting the earth from harsh ultraviolet rays, lower level ozone (or photochemical smog) contributes to a variety of health problems. Additional problems are posed by fine particles in the atmosphere. Under certain conditions in the air, oxides of nitrogen can undergo additional reactions resulting in fine particles or contribute to acids in aerosol droplets. EPA has set health-based National Ambient Air Quality Standards (NAAQS) for NO_2, for ozone, and for fine particulates (which have many sources in addition to nitrogen oxides). Acidic precipitation is also affected by nitrogen oxide emissions, and is the focus of another EPA program. Recent publications from those programs and NAAQS reviews should be consulted for details on the human health effects of nitrogen. The Great Waters program evaluates the reductions in nitrogen compounds that these programs have achieved and have proposed, and considers the net effects on waterbodies, but does not analyze direct human health effects from inhalation.

II.E Other Effects

In addition to adverse effects on human and ecological health, atmospheric deposition of the pollutants of concern may contribute to other adverse effects including environmental justice concerns (e.g., effects on subsistence fishermen), commercial and recreational fishing losses, and other recreational losses. This section provides a very brief overview of these other potential effects. This section does not attempt to establish a link between these effects and atmospheric deposition of the pollutants of concern to the Great Waters.

Environmental Justice Concerns

As introduced in the First Report to Congress, an important environmental justice issue in the Great Waters is whether certain groups of people may have higher exposures to the pollutants of concern than the general population, and therefore potentially greater risks for adverse health effects. One potential effect, cancer, is discussed in the box below (note that noncancer effects may be as or more important).

Subpopulations with Higher Lifetime Cancer Risks

In developing the Great Lakes Water Quality Guidance (GLWQG), EPA estimated baseline human health (cancer and noncancer) risks for certain subpopulations in each of the Great Lakes. Baseline health risks were based on fish tissue concentrations of chlordane, DDT, dieldrin, hexachlorobenzene, mercury, PCBs, 2,3,7,8-TCDD, and toxaphene. Table II-11 presents the low and high estimated lifetime cancer risks, as well as EPA's "acceptable" range of lifetime cancer risk for human health. As shown in the table, the risks to the subpopulations are well above the accepted risk range.

TABLE II-11
Lifetime Cancer Risks in Various Great Lakes Subpopulations Versus EPA's Appropriate Range of Risk to Human Health[a]

Subpopulations	Low	High
Native Americans (subsistence anglers)	18 in 10,000	370 in 10,000
Low income minority sport anglers	25 in 10,000	120 in 10,000
Other sport anglers	9.7 in 10,000	450 in 10,000
EPA's Acceptable Range of Lifetime Cancer Risk for General Population[b]	0.01 in 10,000 to 1 in 10,000	

[a] Cancer risks were driven by fish tissue PCB concentrations which were lowest in Lake Superior and highest in Lake Michigan. Therefore, low end of range represents Lake Superior and high end represents Lake Michigan.
[b] EPA's acceptable range of lifetime cancer risk is based on the National Oil and Hazardous Substances Pollution Contingency Plan (NCP) [40 CFR Part 300].

Sources: U.S. EPA 1995a, 1995h.

Some populations are at higher risk because they may be more vulnerable to effects (e.g., children). Other populations may be more highly exposed because they consume larger quantities of Great Waters fish than the general population (e.g., for subsistence reasons); these populations include Native Americans, the urban poor, and sport anglers. For example, researchers found a relationship between fish consumption levels and socioeconomic characteristics in a study of Michigan licensed sport anglers. A combination of minority status and relatively low annual income (less than $25,000) was correlated with higher levels of fish consumption (West et al. 1993). Higher fish consumption rates have been correlated with higher levels of contaminants in the blood:

- High levels of dioxins and furans were found in frequent consumers of fish living near the Baltic Sea in Sweden. Individuals with the highest percentage of fish in their diet, specifically fishermen and fishing industry workers, had blood levels that were approximately three times that of non-fish consumers (Svensson et al. 1991).

- On the north shore of the Gulf of the St. Lawrence River, fishermen who consumed large amounts of seafood had elevated levels of PCBs in the blood. The blood levels among the highly exposed fisherman were 20 times higher than controls (Dewailly et al. 1994).

As indicated in Section II.A, fish consumption advisories are designed to take into account the fact that some populations are potentially at a greater risk for exposure and potential effects than the general population. Recent studies indicate that some high-risk populations are changing their fish consumption and preparation habits in response to fish consumption advisories. A survey of 8,000 sport anglers in the Great Lakes states found that 36 percent of the respondents had changed their fish consumption behaviors, including modifying fish cleaning and preparation methods and eating less Great Lakes fish (Connelly and Knuth 1993). Another study found that pregnant women of the Mohawk nation had substantially reduced their fish consumption (Fitzgerald et al. 1993).

While fish advisories are designed to the reduce the harmful health effects from eating contaminated fish, they may have negative cultural, societal, and economic impacts. A few of these factors are described briefly below (adapted from U.S. EPA 1996a):

- *Nutritional Value*. Fish are known to be high in important nutrients such as protein and are generally low in fat. By heeding advisories to limit or avoid fish consumption, people may reduce their intake of an important food source without supplementing their diet with other nutritional foods or vitamin supplements. This is especially true for poor subsistence fishers; after fish consumption advisories are issued, these people may not be able to afford other means for eating a well-balanced diet.

- *Health Benefits of Fish Consumption*. A diet rich in fish may have some important health benefits, as demonstrated by studies that have compared diets high in fish with traditional western diets (summarized in U.S. EPA 1996a). Examples of possible health benefits from diets with a high proportion of fish are reduced risk for cardiovascular disease and certain cancers. Therefore, the health risks from not eating fish may outweigh the potential health risks from eating contaminated fish.

- *Traditional Activities*. Eating and catching fish are important cultural activities for many subgroups. For example, for centuries some Native American tribes have built cultural traditions around spearing fish and sharing the catch (U.S. EPA 1992). Tribes that live near large waterbodies often use certain fish species to symbolize characteristics or ideas. For sport anglers, fishing and eating their catch are important social activities. Issuing fish advisories can impose on these important cultural activities, greatly affecting a subgroups' traditional values.

- *Dietary Patterns*. Subgroups such as Native American and Asian American communities have long-standing traditions of eating fish and place importance on celebratory meals highlighted by fish. People may not want to substitute different ingredients or cook in different ways. In addition, the potential substitutes for fish may be more unhealthy and expensive.

- *Fishing and Tourism Industries*. As discussed below, fishing advisories may negatively affect the fishing and tourism industries.

An on-going Canadian study established as a First Nations/Health Canada partnership is assessing the extent of exposure of Native people living in the Great Lakes basin to bioaccumulative pollutants and the associated risk to their health and well-being. The project, Effects on Aboriginals from the Great Lakes Environment (EAGLE), began in September 1990 and is expected to be completed in 1997. EAGLE is a community-based epidemiological project involving the approximately 100,000 First Nations people living in 63 aboriginal communities in the Great Lakes Basin. The project builds on earlier studies and is examining exposure in both adults and children, socio-economic effects, and the impacts on traditional ways of life, culture, and values (Manno et al. 1995).

Commercial and Recreational Fishing Losses

> The Arkansas Game and Fish Commission has estimated a loss of fishing expenditures due to mercury fish consumption advisories of over $5 million dollars from 1991 to 1992.
>
> Source: Armstrong 1994.

Bans and advisories on commercial fishing due to pollution can cause economic and social losses to owners and employees of commercial fishing enterprises. In addition, bans and advisories on recreational fishing have potentially far-reaching effects on the U.S. economy; expenditures on recreational fishing stimulate the economy, provide jobs in the industry, and generate state and federal taxes (Fedler and Nickum 1992). In 1991, more than 35.6 million Americans over 16 years of age spent a total of $24 billion on sport fishing, averaging 13 trips each and 14 days a year (U.S. FWS 1993). As shown in the sidebar, losses due to fish consumption advisories can be significant.

Other Recreational Losses

In addition to recreational fishing, recreational hunting (e.g., for food or fur) may be negatively impacted by exposure of wildlife to the pollutants of concern. These impacts include decreases in the populations of hunted species (e.g., stemming from adverse reproductive effects) and increased health risks to the hunters who consume their kill. Poor water quality (and/or public perception of poor water quality) may negatively affect recreational uses other than fishing and hunting, such as boating, swimming, and visiting parks. Decreased recreational use also may affect the related tourism economies; the annual outdoor recreation/wildlife industry in Great Lakes states generates approximately $12 billion (NWF 1993). An example of efforts to study the impact of recreational losses in one of the Great Waters is presented in the sidebar.

EPA is currently funding a project that will assess the value of certain recreational services (e.g., bird watching, windsurfing, fishing) to the Corpus Christi Bay National Estuary Program. The objective of this project is to assist in prioritizing management actions for this estuary. Findings from the project will be used, in part, to provide a framework for assessing both economic and natural resource value losses due to negative impacts associated with excess nitrogen loadings to Corpus Christi Bay and other estuaries.

CHAPTER III
ATMOSPHERIC TRANSPORT AND DEPOSITION PROCESSES

Section 112(m) of the Clean Air Act (CAA) directs EPA, in cooperation with the National Oceanic and Atmospheric Administration (NOAA), to assess the extent of atmospheric deposition of hazardous air pollutants to the Great Waters. As part of this assessment, EPA is directed to, among other things, monitor atmospheric deposition of pollutants, investigate pollutant sources and deposition rates, and conduct research to improve monitoring methods. Accomplishing these tasks requires an understanding of the processes by which the pollutants of concern are transported from their emission sources and deposited to the Great Waters. Researchers use mathematical models of atmospheric transport and deposition of pollutants to analyze the movement of pollutants in the environment, to develop relationships between sources and receptors of pollutants, and to evaluate prospective control strategies.

This chapter introduces some basic concepts and scientific terms that are relevant to atmospheric transport and deposition of pollutants, and that are used throughout this report. It also discusses general types of transport and deposition models, focusing on modeling achievements of the most recent years and on efforts by EPA and NOAA plus other researchers, mostly in academia. Further discussion of the uses of a few of these models is included in Chapter IV.

There is currently a general understanding of the major factors that affect the transport of air pollutants between their sources and receptors, as well as how these factors interrelate. The development of new models and refinement of existing ones have been advanced in recent years, as shown in this chapter. Research on the characterization of sources, processes, and parameters has continued in parallel with monitoring efforts. The Great Waters have been and will continue to be the focus of many of the investigations presented in this chapter.

Despite recent advances in our understanding of transport and deposition of contaminants in the atmosphere, there is still a paucity of data with which to calibrate models (measurement of dry deposition and source inventories are typical examples). Further analysis is also required for some transport and deposition phenomena, such as the importance of environmental cycling of contaminants emitted and deposited in the past.

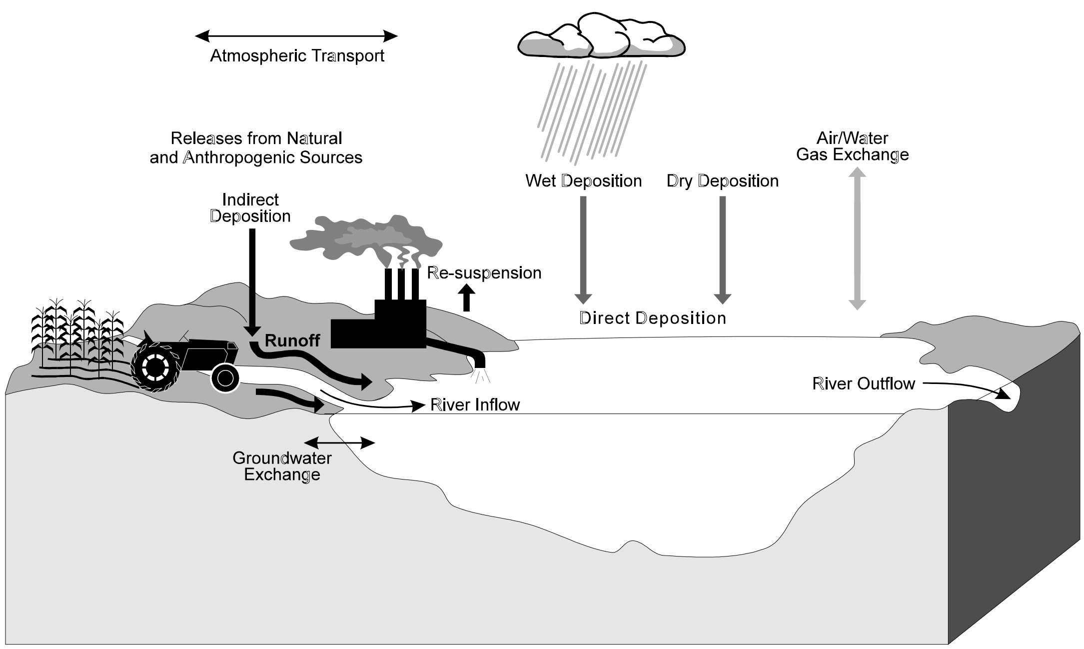
This chapter is divided into three sections. Section III.A discusses some key terms, such as deposition and environmental cycling. Section III.B presents a brief description of general types of transport and deposition models. Section III.C compares some models that have been recently used in Great Waters studies, and discusses modeling and data limitations.

III.A Atmospheric Deposition and Environmental Cycling

Long-range atmospheric transport and deposition of pollutants have been widely acknowledged to make a significant contribution to contaminant inputs to surface waters, including the Great Waters. Atmospheric deposition refers to the removal of pollutants (following transport) from the air to soil, water, and other surfaces. Deposition may occur directly to the water surface and/or indirectly to the land surface in the watershed, with subsequent runoff from rainfall carrying contaminants to the waterbody. It is important to recognize that both local and distant air emission sources may contribute to the pollutant loading at a given location and time. Also, pollutants may be emitted from a combination of point, mobile, or area sources; or even from resuspension in the air of previously deposited material.

There are three major processes of direct atmospheric deposition to natural waterbodies: wet deposition, dry deposition, and gas exchange across the air-water interface. A schematic of these processes is presented in Figure III-1. In addition to these processes, cycling of semi-volatile compounds (e.g., polychlorinated biphenyls (PCBs)) and persistent compounds among environmental media (e.g., air, water, sediment) can be an important input of pollutants in waterbodies.

FIGURE III-1
Atmospheric Deposition Processes



Monitoring and assessment of these processes provide important information on the atmospheric contribution of a given pollutant to a specific site, as well as extent of deposition of the pollutant. Hence, monitoring at Great Waters study sites requires meteorological measurements and measures of contaminant concentrations very near to the site and, if possible, over the surface of the waterbody itself. Understanding of these processes allows researchers to evaluate quantitatively the long-term distribution of a pollutant in an aquatic system. EPA sponsors research on atmospheric deposition so that future monitoring can become less research-oriented and more focused on evaluating trends in atmospheric loading of pollutants and determining the effects resulting from efforts to reduce emissions.

Wet Deposition

Wet deposition (or in more general terms, removal via precipitation) refers to the incorporation of both gases and particles into cloud droplets and into "hydrometeors" (e.g., snow, sleet, and rain) where they are carried to the Earth's surface in precipitation. Pollutants may be removed from air by wet deposition through three main mechanisms:

(1) minute pollutant-bearing particles can serve as cloud condensation nuclei and become naturally embedded into water droplets during cloud formation; (2) particles may be incorporated into falling hydrometeors by collisions; or (3) gaseous pollutants may be dissolved into cloud droplets and falling rain. Cloud and precipitation droplets, therefore, may contain pollutants in both particulate and dissolved forms. The removal of gases by wet deposition is dependent on their solubilities in the elements of precipitation. Wet deposition is essentially an intermittent process, although if precipitation events are frequent or long-lasting, wet deposition may be the major pathway for deposition of air pollutants, specifically for totally particulate species (e.g., metals other than mercury). For semi-volatile compounds, air-water gas exchange appears to be a dominant factor (Hoff et al. 1996).

To get useful and quantitative measurements of wet deposition, well-prepared protocols and quality assurance approaches are designed and used, with careful handling of field equipment, samples, and subsequent chemical analyses. For example, wet deposition rates are determined using collectors that are designed to open only during precipitation events. Work must be designed and carried out so as to minimize measurement problems with the samples that can arise from chemical or biological activity, evaporation of gaseous pollutants, or possible contamination during storage and handling. The concentrations of most of the pollutants of concern in any one precipitation event may be small, so rather advanced field and chemical-analytical methods are needed for quantitatively assessing the concentrations. Though measurements must be taken of short-term events in which the concentrations may be small, it is important to track the concentrations of the pollutants of concern deposited over time because of their persistence and tendency to bioaccumulate. Thus, the kind of monitoring used in the Great Waters program involves research into new and more accurate monitoring techniques and must be carefully designed to focus efforts on well-located sites, protocols, and methods.

Dry Deposition

Dry deposition refers to removal from the air of pollutants (bound to particles or in the gaseous form) to the land or water surface in the absence of precipitation. Dry deposition is essentially a continuous process and often represents a substantial removal of the pollutants from the atmosphere. The pollutants reach the surface by the turbulent movements of the air or, for large particles, through gravitational settling. Dry deposition of particulates with high deposition velocities is an important contribution to pollutant loadings to waterbodies located near cities. For large open waterbodies, air-water gas exchange appears to contribute more than dry particle deposition (Hoff et al. 1996). A review of models of dry deposition to water is presented by Zufall and Davidson (1997).

Research on Dry Deposition

In 1994, a research program was established among the EPA, the University of Michigan, the Illinois Institute of Technology, Carnegie-Mellon University, and Oak Ridge National Laboratory to develop and test new techniques for measuring dry deposition of mercury and other trace elements to natural water surfaces. The data will be used to develop new mathematical models for predicting dry deposition onto water surfaces under a variety of atmospheric and surface conditions. Measurement methodologies have been developed and tested, and modeling is underway. Field studies in 1996-1997 are planned to gather basic data, and to compare with model predictions.

Dry deposition is measured from pollutant accumulation on artificial surfaces (e.g., a plexiglass plate). Alternatively, atmospheric pollutant concentrations and their mass-size distribution can be combined with micrometeorology data to model dry deposition rates. The major difficulty in measuring dry deposition is that there is no method that can be routinely used because the flux density is so low and because the exchange rate is usually governed by details of the surface that cannot be reproduced in artificial devices. The difficulties of measuring dry deposition have motivated new research on measurement methodologies and modeling of dry deposition under the Great Waters program (see sidebar on the previous page).

Gas Exchange Across the Air-Water Interface

In addition to wet and dry deposition of pollutants, gaseous pollutants may be directly exchanged between air and water (i.e., transferred in either direction across the air-water interface). The gaseous exchange of organic compounds at the air-water interface is an important phenomenon in the balance of pollutants occurring in air and water (Eisenreich et al. 1997), and extensive North American waterbodies, such as the Great Lakes and Chesapeake Bay, provide large surface areas for this exchange to occur. Analogous gas exchange phenomena occur between plants and air, and land and air.

Gas exchange is a two-way process involving both ***gas absorption*** or invasion (air to water) and ***volatilization*** or evasion (water to air) across an air-water interface of a volatile chemical (usually in gaseous form under every-day conditions) or a semi-volatile chemical (e.g., polycyclic aromatic hydrocarbons (PAHs), PCBs). The direction of gas movement (from air to water or from water to air) depends on the fugacity difference (i.e., the relation of the Henry's law scaled air concentration to the water concentration). The direction of gas exchange will tend to reduce this difference and move toward a near-equilibrium condition. The direction and magnitude of gas transfer is a function of the chemical concentrations in air and water, wind speed, temperatures in air and water, waves (height, frequency), physical and chemical properties of the pollutant (e.g., molecular weight, vapor pressure, Henry's Law constant, solubility), and in some cases characteristics of the water (e.g., pH for acidic and basic species, and salinity in estuaries).

Gas absorption and volatilization occur simultaneously, even when near-equilibrium has been achieved. Taken together, volatilization and gas absorption contribute to the ***net flux*** (the difference between the amount of pollutant invading and the amount evading) or effective movement of a chemical across the air-water interface. Net flux may be expressed as the mass of gas moving across a unit of area over a unit of time (e.g., 8 ng per m^2 of water surface per day). To achieve quantitative estimates for the Great Waters of net deposition of pollutants of concern, many of which are semi-volatile, such physical and chemical processes must be quantified so they may be correctly used in models. Some earlier work on pollutant loading made simplifying assumptions of "one-way" flux or deposition without quantifying gas exchanges, but recent work shows the need for more complete representation of the natural processes for each pollutant. Furthermore, it is important in some cases to determine both absorption and volatilization, instead of net flux alone. Even under conditions close to air-water equilibrium, with small net flux, the absorption and volatilization may be quite large, making gas exchange a key factor in the analysis of pollutant movements (see Table IV-2 in Section IV.A on the Great Lakes).

There are two models commonly used to describe gas exchange at the air-water interface. The Stagnant Two-film Model (Liss and Slater 1974; Whitman 1923) is used to estimate gas exchange at low wind speeds, which results in an essentially stagnant boundary layer just above the water. However, as higher wind speeds generate more turbulence in the boundary layers of both air and water, turbulent eddies bring small parcels of water to the surface, where they begin to equilibrate with the atmosphere, and more rapid gas exchange of chemicals occurs. To characterize this situation, the Surface Renewal Model may be used, but the choice of model for a given situation is not always clear-cut. The air-water gas exchange models continue to be improved to incorporate new factors such as effect of bubbles, breaking waves, and surface films. For a review of these models, readers should refer to Theofannous (1984) and Eisenreich et al. (1997).

Environmental Cycling of Semi-Volatile Compounds

The exchange and cycling of gases between air, water, and soil is especially important for the pollutants of concern that are characterized by chemists as "semi-volatile" in nature. These semi-volatile pollutants coexist in the atmosphere in both the gas and particle phases, and can revolatilize into the air after deposition (though not as readily as volatile compounds such as benzene or vinyl chloride). Semi-volatile organic compounds include PAHs, PCBs, and several pesticides (e.g., hexachlorobenzene, α-HCH, and lindane), as well as the metallic form of mercury and many of the other 188 toxic pollutants listed as hazardous air pollutant (HAPs) in section 112(b) of the CAA. Because these compounds possess very low vapor pressures and water solubilities, they are distributed between the gaseous and particulate phase both in the atmosphere and in the water column, and their distribution among air, water, soil, and vegetation is very complex. This makes tracking or modeling movements of semi-volatile compounds very difficult. Each pollutant has particular chemical-physical attributes affecting phase distribution, so quantitative assessment requires that each pollutant be well understood. Some of the pollutants of concern can be chemically persistent, maintaining chemical identity (not being broken down) as they move among physical phases and, in many cases, into biological tissues. Some other pollutants (such as PAHs) may change to compounds that are bioaccumulative and more toxic than the parent compounds. Other groups of compounds may separate into individual components due to interactions and relative movement among solids and liquids in the environment (Wania and Mackay 1993).

Once released to the environment, persistent semi-volatile compounds may repeatedly cycle between the atmosphere, land, and waterbodies. This cycle can extend over long time periods, resulting in transport of the compounds for long distances. Long-distance transport with repeated deposition to land or water and then revolatilization to the atmosphere has been shown to occur in response to seasonal temperature changes, among other factors (Wania and Mackay 1996). Other factors that influence the extent and duration of this cycling include volatility and persistence of the substance; molecular weight; concentrations and temperatures in air, soil, and water; and atmospheric circulation, pressure, and weather conditions. As the seasons change, the behaviors of atmospheric contaminants change relative to their location in physical media; therefore, sampling work and modeling calculations must be adjusted, for each compound, to correctly estimate their presence and impacts. Warmer conditions on seasonal and global scales generally favor net movement into the atmosphere. Often redeposition takes place in areas of colder atmospheric temperatures (Wallace et al. 1996; Wania and Mackay 1993, 1996). The modeling of chemical fate and concentrations of semi-volatile pollutants over very large areas is challenging, and lack of data on pollutant source and release makes the validation of existing models difficult (Wania and Mackay 1996). Also, the lack of definitive methods to measure the concentration of the mix of gases, particles, and liquid droplets that constitutes some of the organic semi-volatile pollutants being deposited poses significant challenges for validation of models.

III.B Atmospheric Transport and Deposition Models

The emission, transport, transformation, and deposition of pollutants to the Great Waters is a complex series of processes involving different pollutants that have different behaviors in air and water systems, over very large geographic areas. Therefore, numerous models, as well as input parameters for these models, have been and continue to be developed or evaluated for estimating the atmospheric transport and deposition processes for the various pollutants associated with the Great Waters.

Atmospheric transport and deposition models are used to:

- Predict the direction and distances pollutants will travel in the environment;
- Test hypotheses about characterizations of atmospheric transformations and removal;
- Assist in designing monitoring networks for efficiency and specific analyses, and in placing a limited number of monitors effectively;
- Provide calculated estimates to fill spatial and temporal gaps in monitoring networks, to provide a smooth or coherent picture for analyses;
- Develop relationships between sources and receptors of pollutants; and
- Evaluate prospective pollutant control strategies.

This section discusses different types of models related to atmospheric transport and deposition processes. Results from application of some models presented in this section are described in Chapter IV. The models presented in this section can be classified in three categories: mass balance models, source apportionment techniques, and air quality simulation models.

Mass balance models analyze all ways that pollutants can enter and exit a waterbody, and their corresponding amounts over a period of time (commonly referred to as the pollutant loading). In other words, mass balance models consider the mass (or weight) of a pollutant that is exchanged across interfaces between air, water, land, and sediments as inputs and outputs, to assess the relative loadings of a pollutant into a waterbody by different pathways.

Source apportionment techniques attempt to link sources and receptors of pollutants of interest. Primary source apportionment techniques include *dispersion models, receptor models, and hybrid models*. Dispersion models trace pollutants from their sources to the air at given locations (e.g., a waterbody). Receptor models trace pollutants in the air at a given location back to particular source types. Hybrid models are similar to receptor models, but also incorporate meteorological data, and work from both the source end and the receptor end of the pollutant transport analysis. One important uncertainty related to source apportionment techniques is the lack of complete and reliable input data, such as the composition and emissions of some pollutant sources. In addition, source apportionment techniques cannot be applied well for air pollutants that are widespread in the environment, travel over long distances, and/or are emitted in large quantities from natural sources or broad area sources. A review of source apportionment techniques for organic compounds, with special emphasis on the Great Waters, is presented by Keeler et al. (1993).

Mass Balance Models. Estimate inputs and outputs of a pollutant to a waterbody (i.e., total amounts of a given pollutant that enter and exit a water body by each of the various pathways).

Source Apportionment Techniques. Estimate the relative contribution of different sources to air pollutant levels at a specific receptor site (e.g., a particular air mass over Lake Michigan on a particular day).

Air Quality Simulation Models. Use extensive source emission inventories, meteorological data, and algorithms to simulate processes such as dispersion of pollutants in the atmosphere, transformation of compounds, and deposition.

Air Quality Simulation Models (AQSMs) are used to characterize both the transport and deposition of pollutants. As input data, these models use extensive source emission inventories and meteorological data. AQSMs include algorithms to simulate processes such as dispersion of pollutants in the atmosphere, transformation of compounds, and deposition. The models' results typically include air concentrations and deposition rates of pollutants over a given area for a specified period of time. Although comprehensive, AQSMs are limited by the quality of input data, the computational difficulties of their algorithms, and the modeling of some processes (e.g., air-surface exchange).

The remainder of this section presents additional information on specific applications of the model types mentioned above, especially those that have been recently applied to Great Waters studies. The following subsections focus on mass balance models, receptor models, and AQSMs.

Mass Balance Models

A mass balance model provides the essential framework for determining the relative contribution of pollutant loadings from various mechanisms of input (e.g., direct discharge, river input, atmospheric deposition) and output (e.g., sedimentation, volatilization, outflow) to and from a waterbody. Mass balance models are also helpful to relate concentration measurements to pollutant mass fluxes between different media (air, waterbodies, land surface) and to mass of a contaminant in different environmental "pools" (e.g., a waterbody, a land region). As introduced in the First Report to Congress, when reliable information is available for contributions from the various sources, mass balance models may be used to estimate the importance of atmospheric deposition (or any other mechanism) in causing contamination of a waterbody. Mass balance models usually are good at recognizing sizable pollutant sources and receptors, but often lack the resolution needed to deal with multiple smaller sources that by themselves are not significant, but added together could be important in some situations.

Mass balance studies have provided insights on atmospheric deposition issues relevant to the Great Waters. A review of studies on relative atmospheric loadings of toxic contaminants and nitrogen to the Great Waters is presented by Baker et al. (1993). These studies have provided quantitative estimates indicating that: (1) atmospheric deposition can be the main contributor of toxic chemical contamination and nitrogen enrichment to the Great Waters, although uncertainties still exist; (2) the importance of atmospheric load for a specific pollutant in a given waterbody depends on characteristics of the waterbody, chemical properties, and source locations; and (3) chemicals may cycle between soil, air, water, and biota for many years.

The First Report to Congress presented mass balance case studies for some Great Waters pollutants of concern, such as PCBs in Lake Superior, mercury in lakes in Wisconsin, and nitrogen in the coastal waters of several Atlantic states. Considerable research continues on the development and use of mass balance models for the pollutants of concern in the Great Waters. The Lake Michigan Mass Balance Study, an EPA-sponsored research project, is responsible for a comprehensive sampling effort in Lake Michigan, including measurements of pollutants such as PCBs, trans-nonachlor (a component of technical grade chlordane), mercury, and atrazine in the atmosphere, tributaries, open lake water, sediments, and food chain (fish tissue). Samples collected over a two-year period (1994-1995) for modeling will be used to improve understanding of key environmental processes that govern cycling and bioavailability of contaminants within the Lake Michigan ecosystem. The approach modifies the classic mass balance within a closed system to consider inputs from transport, including long-range transport. Analysis of some selected modeling runs is expected in 1998. A more detailed discussion of this mass balance project is described in Section IV.A.

Receptor Models

Receptor models, which are one type of source apportionment technique, trace pollutants in the air at various locations (such as over a waterbody) back to particular source types in order to estimate the contribution to pollutant levels from a group of sources with similar emissions. This type of model does not use the detailed meteorological data or extensive emission inventories used in air quality simulation models. Receptor models assume that chemicals of concern are affected in the same way by all of the processes involved in pollutant transport and dispersion. This is a particularly useful assumption, but it presents some difficulties when clouds are present, when precipitation occurs, or when extensive chemical transformations of a pollutant are known to occur. A limitation of the receptor models is the lack of adequate "source profile" data, which allow air pollution to be linked to a particular source type. Source profiles refer to "signatures" or "fingerprints" of emissions from a

Receptor Model for Great Waters

A project on atmospheric deposition in the Great Waters, entitled Atmospheric Exchange Over Lakes and Oceans (AEOLOS), was started in 1993 by EPA and scientists from the Universities of Minnesota, Michigan, Maryland, Delaware, and the Illinois Institute of Technology. The objectives of this 4-year research program are to determine: (1) the dry depositional fluxes of critical urban contaminants to northern Chesapeake Bay off Baltimore and southern Lake Michigan off Chicago; (2) the contributions of urban source categories to measured atmospheric concentrations and deposition; and (3) the air-water exchange of contaminants and their partitioning into aquatic phases. The contaminants being studied include mercury, trace metals, PAHs, and PCBs. Techniques involve using all three modeling approaches described below -- CMB, PCA, and trajectory analyses. Research is expected to be published in 1998.

type or category of sources; these profiles are determined through samples taken from the actual emissions (i.e., from the "smokestacks") followed by analyses comparing the chemical signature to those of other categories of sources. Despite the limitations of receptor models, these models provide useful insights into contaminant transport to the Great Waters (see sidebar on the previous page).

Some receptor model types include:

- *Chemical mass balance (CMB)*: The CMB model assumes that emission characteristics (i.e., chemical and elemental composition, physical size, morphology) of various source types are sufficiently different from one another that their contributions to a receptor may be identified by measuring the characteristics in samples collected at the site. The observed concentration pattern of an ambient sample at the receptor site is equated to a linear combination of the appropriate pollutant source patterns, each weighted by an unknown source strength term. The primary application of the CMB model has been to urban areas such as Chicago and Baltimore. CMB models assume composition of all contributing sources are known, and when this is not the case the uses of the model are limited.

- *Principal component analysis (PCA)*: The objective of PCA is to use mathematical analyses to find a minimum number of factors, or source categories, that explain most of the variance in a set of measurements from a receptor site, instead of using all sources as in the CMB models. The number of statistically significant factors is usually found to be six or less. PCA is often limited because it lacks fine resolution of contribution from various distant sources. An advantage of PCA is that ancillary measurements (e.g., wind speed, wind direction, relative humidity) may be incorporated into the analysis along with pollutant concentrations.

- *Trajectory clustering*: In these models, a back trajectory is computed using wind data, and the spatial probability of an air "parcel" reaching a particular receptor site at a particular time is calculated. Under different meteorological conditions, all potential trajectories and concurrent pollutant measurements are grouped into a more manageable set of source clusters and regions. A variant of this approach is being used to assess mercury deposition in the Great Lakes basin (see sidebar).

Trajectory Clustering Technique in the Great Lakes

To determine the sources of mercury deposition to the Great Lakes Basin, a regional network of 10 monitoring sites was established in 1993 by EPA and the University of Michigan to measure atmospheric mercury over several years. The sampling will continue into 1997. Data will be analyzed, using an improved trajectory clustering technique, to determine the sources and source areas most responsible for mercury deposition to the Great Lakes.

Source: Burke and Keeler 1995.

Air Quality Simulation Models

Air quality simulation models (AQSMs) are used to characterize the emission, transport, and deposition of hazardous air pollutants over large geographic areas. These models incorporate fairly extensive source emission inventories and meteorological data bases (e.g., wind fields, temperature, mixing height), and apply the collected data to simulated processes such as dispersion, transformation, and deposition. The models are run to generate estimates of pollutant concentrations and deposition rates over a spatial and temporal pattern. AQSMs are based on two approaches. In one approach, characteristics or properties of air are assigned to fixed points in space at a given time. The second approach is based on a two- or three-dimensional grid system for the geographical pattern of interest, and all the fundamental processes (e.g., emissions, chemical and physical transformations, deposition) of discrete air parcels are considered to occur within the individual grids or boxes.

The mathematical relationships between emissions and concentration (or deposition) are typically nonlinear, due to the influences of atmospheric transport, chemical and physical transformation, and deposition processes. AQSMs attempt to model the nonlinear physical and chemical processes influencing atmospheric concentrations and deposition. AQSMs may be useful tools in providing the analytical framework required to predict the environmental impacts of proposed emission control programs and, consequently, performing both scientific and regulatory assessments.

This section describes two of the various AQSMs that have been developed and peer-reviewed in recent years, the Regional Lagrangian Model of Air Pollution (RELMAP) and the Regional Acid Deposition Model (RADM). These two models (among others presented in Section III.C) have been used in Great Waters studies, and their results are presented in Chapter IV. Another model being developed for application to atmospheric deposition in the Great Waters is discussed in the sidebar.

A Model for Assessing Atmospheric Deposition to the Great Waters

EPA has recently developed a new modeling tool for the assessment of atmospheric deposition of pollutants to the Great Waters. The Regulatory Modeling System for Aerosols and Deposition (REMSAD) is a work station-based Eulerian model intended for use in assessing the impacts of regulatory activities, such as the MACT standards, on loadings of pollutants of concern to the Great Waters. REMSAD is currently capable of simulating short-, medium-, and long-range transport and deposition of cadmium, dioxins, mercury, and POM. Nitrate deposition distributions have been produced through REMSAD simulation, but have not yet been compared to other models such as RADM. Other pollutants, including other toxics, be incorporated in future work. Initial model demonstration and evaluation will be completed during 1997. The model is currently available on the OAQPS Support Center for Regulatory Air Models (SCRAM) bulletin board.

RELMAP is used to simulate the emission, transport, and diffusion of pollutants, their chemical transformations, and wet and dry deposition. The model was originally designed for sulfur analysis (the User's Guide is presented in Eder et al. (1986)). It has also been applied to mercury and other toxic metals (Bullock et al. 1997; Clark et al. 1992), among others. For example, the goal of one study was to determine the extent of mercury emissions to air in the United States over an entire year, the deposition to U.S. soil and waterbodies, and the contribution by source category to the total amount of mercury emitted and deposited within the United States (Bullock et al. 1997). Section III.C presents relevant applications and limitations of this model.

RADM has been developed over the last ten years under the National Acid Precipitation Assessment Program (NAPAP) to address policy and technical issues associated with acidic deposition (Chang et al. 1990; Dennis et al. 1990). The version of RADM used for NAPAP models an area east of Central Texas and south of James Bay, Canada, to the southern tip of Florida. This area is divided in a grid, and for each grid cell the model considers pollutant emissions, transport in and out of the cell, turbulent motion in the atmosphere, chemical reactions that produce or deplete the chemical, vertical transport by clouds, and removal by dry deposition. RADM is designed to model 140 chemical reactions among 60 pollutants, 40 of which are organic compounds. A feature of RADM is the simultaneous modeling of sulfur and nitrogen deposition. This is an important consideration because the amount of sulfur dioxide (SO_2) present in the atmosphere affects the formation of both sulfates and nitrates and thus, the amount and spatial distribution of nitrogen deposited back to waterbodies. RADM is also useful for analyzing long-range transport issues, but it is very complex computationally. Recently, it has been applied to the study of nitrogen deposition in the Chesapeake Bay watershed (Dennis 1997).[9] The results of this study are presented in Section IV.C. The limitations of this study, as well as more background information on RADM, are presented in Dennis (1997).

AQSMs are limited by the quality of the algorithms used to simulate various processes affecting pollutants of interest (e.g., chemical transformation, deposition), quality of input data (e.g., emissions, meteorology), and lack or inadequacy of modeling of certain processes (e.g., air/water gas exchange). The quantity and quality of available input data is an important limiting factor in the application of AQSMs, especially for atmospheric pollutants, such as many pesticides and PCBs, that have relatively poorly developed emission inventories, or for which re-emission and environmental cycling are significant.

Efforts sponsored by the Great Waters program are underway to improve the quality of emission estimates for the HAPs, which historically have been inventoried only in a few places and for short time intervals. In one effort, the eight states that border the Great Lakes have worked together, with EPA, to develop an approved protocol for a coordinated emission inventory of 49 HAPs, including the Great Waters pollutants of concern other than pesticides. The inventories for point sources and area sources, as well as mobile sources, should be completed by 1997-1998. The data are stored in a regional data base system (the Regional Air Pollutant Inventory Development System, or RAPIDS) developed for this project. In another effort, EPA is in the process of developing national inventories of sources and emissions for seven specific HAPs (hexachlorobenzene, alkylated lead compounds, PCBs, POM, mercury, 2,3,7,8-TCDD, and 2,3,7,8-TCDF) in response to the mandate in section 112(c)(6) of the CAA.

III.C Comparing Models Used in Great Waters Studies

Several numerical atmospheric transport and deposition models or modeling strategies have been and continue to be developed and used for understanding deposition of pollutants to the Great Waters. Models have many roles in EPA's atmospheric programs and are widely used to link emissions data, meteorology, receptor sites (e.g., people, or lakes, exposed to pollutants), and monitoring of the ambient air (in cities, or over lakes for the Great Waters). New approaches to modeling have been needed to deal with the particular complexities of the issues in Great Waters studies. To discuss these models and their applications to the Great Waters with the general scientific community, the EPA Great Waters program co-sponsored a session at the 15th

[9] Work by Dennis (1997) models nitrate deposition only; however, ammonium and organic nitrogen deposition may also be quantitatively important to the Bay.

annual meeting of the Society of Environmental Toxicology and Chemistry (SETAC) in 1994 on "Atmospheric Deposition of Nutrients & Contaminants." Following that meeting, the presenters and other scientists prepared written chapters for a SETAC Special Publication: *Atmospheric Deposition of Contaminants to The Great Lakes and Coastal Waters*, J.E. Baker, editor (Baker 1997). This section briefly describes the main applications and limitations of the models presented in the SETAC session. This presentation is not intended to be a comprehensive list of all existing models and their uses, as the development of transport and deposition models is a very active research area. The document based on the SETAC session (Baker 1997) provides an in-depth technical reference that supplements the information in this report.

Table III-1 summarizes some of the relevant modeling efforts. These models are further discussed below, with a very brief description of the model, its application to Great Waters studies, and how the model compares with actual monitoring data; further information is presented in Baker (1997).

The first modeling effort listed in Table III-1 is the regional-scale analysis of nitrogen deposition to the Chesapeake Bay watershed (Dennis 1997). The analysis was developed using RADM, described in Section III.B. The overall goals of this study are, first, to define the source region that contributes most of the nitrogen deposition affecting the Chesapeake Bay watershed and, second, to define which source types are most responsible. The modeling results indicate that the range of influence of nitrogen emissions is on the order of 800 kilometers (km) (though this is considered a conservative estimate, given the model bias described in the study). The model indicates that the source region for nitrogen deposition in the Chesapeake Bay watershed is roughly 906,000 km^2, or more than 5.5 times larger than the watershed. Dennis (1997) also uses the model to analyze the spatial distribution of nitrogen deposition by emission sector. For example, the model results indicate that utility emissions tend to be more responsible for nitrogen deposition to the Bay basins themselves, while mobile emissions appear to influence deposition to the mouths of the tributaries and the Bay itself. Additional results of this modeling effort are presented in Section IV.C, although the study does not present any comparison to monitoring data. The study does highlight the importance of additional research on the bias in RADM for nitrogen deposition estimates, processes such as forest or terrestrial retention of nitrogen, and the combined use of air-water models.

The second modeling analysis in Table III-1 looks at wet and dry deposition of semi-volatile organic compounds at a regional scale, with emphasis on the Great Lakes (Ching et al. 1997). The model used is a version of the Regional Particulate Model (RPM), which is itself a modification of RADM that computes the chemical composition and size distribution of the secondary sulfur and nitrogen species (Binkowski and Shankar 1993). Ching et al. (1997) use RPM to analyze the size, chemical composition, and moisture content of airborne particles that serve as sites for condensation and volatilization of semi-volatile organic compounds. Deposition of semi-volatile organic compounds is tracked in the model as proportional to particle deposition. This modeling effort is a step toward the use of regional-scale models to compute air concentrations of these pollutants, and to provide benchmark testing of simpler and computationally less demanding models. Some challenges ahead in this line of modeling include better algorithms and data on air-water gas exchange, the role of clouds as both transporters and chemical transformers, and modeling of resuspension of pollutants from different land uses.

As presented in Table III-1, the numerical modeling of atmospheric mercury by Bullock et al. (1997) uses the model RELMAP (Eder et al. 1986), described in Section III.B, with relatively simple mercury parameterizations. An entire year of transport and deposition of airborne mercury was simulated over the continental United States. The goals of this modeling effort were to analyze the amount of mercury emitted to the air annually over the United States that is deposited back to U.S. soils and waterbodies, the contribution of mercury by source category, and the importance of long-range transport. The RELMAP-simulated annual results agree with the majority of the limited annual deposition and concentration data available around the Great Lakes and in Florida, as well as other areas, usually within a factor of two. Some RELMAP estimates of wet deposition of mercury are somewhat high when compared to actual measurements at those locations. However, the model cannot be well tested over the entire model domain without annual observations in a large number of additional locations. The limitations of this current research effort, including modeling of certain meteorological conditions, aqueous chemistry of mercury, and transport and diffusion modeling are presented in Bullock et al. (1997).

Pirrone and Keeler (1997) propose a hybrid receptor-deposition modeling approach to estimate the dry deposition flux and air-water gas exchange of various HAPs to Lake Michigan. The approach combines modeling of over-water transport of air masses and modeling of deposition and gas exchange. The model parameters were calibrated using data from the Lake Michigan Urban Air Toxics Study (LMUATS) to find both the temporal and spatial variation of critical parameters controlling the transport and deposition of atmospheric contaminants. The results of this study indicate large variations in the parameter values and hence the uncertainty associated with the common practice of using constant parameter values for modeling. The work by Pirrone and Keeler (1997) demonstrates a different approach to estimating surface flux quantities that currently cannot be directly measured with confidence.

TABLE III-1
Summary of Atmospheric Transport and Deposition Models Applied to the Great Waters[a]

Model	Description of Model	Great Waters Related Application	Goodness of Fit[b]	References
Regional Acid Deposition Model (RADM)	Developed under the NAPAP to predict regional changes that may occur as a result of nitrogen and sulfur deposition. The geographic area covered by the model is the eastern U.S. and Canada.	Project quantity and spatial distribution of deposition to the Chesapeake Bay watershed from sources in eastern U.S. and Canada. Estimate fraction that each of 15 subregions in the area contribute to total annual load of atmospherically deposited nitrogen to the Bay watershed and tidal waters.	Comparisons within a factor of 2 for sulfur deposition, and generally within a factor of 2 for nitrogen deposition.	Dennis (1997)
Regional Particulate Model (RPM)	Based on RADM; computes the chemical composition and size distribution of the secondary sulfur and nitrogen species, to identify airborne particles that may serve as sites for condensation or volatilization.	Predict wet and dry deposition of airborne semi-volatile organic toxic compounds to the Great Lakes on a regional scale.	Theoretical only and has not been compared with actual data.	Ching et al. (1997)
Regional Lagrangian Model of Air Pollution (RELMAP)	Simulates concentrations of wet and dry deposition patterns of gaseous pollutants and particulate matter (both fine and coarse), and can generate source-receptor matrices for user-defined regions.	Model deposition of metals including cadmium and lead to Lake Superior; model the emission, transport, and fate of airborne mercury in the U.S., including the Great Lakes and Florida.	Wet deposition results from RELMAP for atmospheric mercury agree with the majority of actual measurements within a factor of 2.	Bullock et al. (1997)
Hybrid receptor-deposition model	Uses backward trajectory calculations and estimates dry deposition and gas exchange flux. Parameters incorporated into the model include transport distance, meteorological conditions, particle size distribution, and water surface roughness.	Estimate deposition of trace metals and semi-volatile organic compounds to Lake Michigan for the Lake Michigan Urban Air Toxics Study.	Experimental model; variation in the model depended on the nature of the chemical species and was ± 3-fold that of values in literature.	Pirrone and Keeler (1997)

[a] Models were presented at 1994 SETAC Annual Meeting and are described in Baker (1997).
[b] "Goodness of fit" refers to how well the deposition estimates from the models correspond to actual measured deposition data.

CHAPTER IV
MAJOR WATERBODIES OF THE GREAT WATERS:
AN OVERVIEW OF PROGRAMS AND EFFORTS ADDRESSING ATMOSPHERIC DEPOSITION

Section 112(m) of the Clean Air Act specifically designates the Great Lakes, Lake Champlain, Chesapeake Bay, and certain other U.S. coastal waters as waterbodies EPA is to consider in identifying and assessing atmospheric deposition of hazardous air pollutants (HAPs) to the Great Waters. Researchers have found that the Great Waters have been affected by metals, pesticides, toxic chemicals, and nutrients that enter the waters through different pathways, including atmospheric deposition.

This chapter presents information currently known about contamination occurring at the individual Great Waters, including problems or issues that challenge each waterbody, followed by discussion of current strategies or efforts to respond to these concerns. Most of the information has been generated from activities occurring at the Great Lakes and Chesapeake Bay. In the Great Lakes, several initiatives have been introduced in recent years, ranging from research projects to gather quantitative estimates of atmospheric loadings to regulatory and voluntary activities promoting reduction of loadings to the waterbody. Efforts at Chesapeake Bay have focused on developing models to improve characterization of nitrogen and toxic contaminant loadings, as part of major reduction strategies. Research is more limited for Lake Champlain and for the other coastal waters. In these waterbodies, smaller-scale investigations have been carried out to study certain pollutants of concern in their respective waterbodies, and in many cases, comprehensive strategic plans have been developed to address contamination issues.

This chapter is organized by sections on each of these major waterbodies of the Great Waters and divided accordingly:

- Section IV.A presents information available on atmospheric deposition of persistent toxic pollutants into the Great Lakes and the many programs to characterize and reduce loadings;
- Section IV.B describes Lake Champlain and current research to assess atmospheric mercury deposition in the basin;
- Section IV.C discusses the deposition of nitrogen and toxic pollutants to Chesapeake Bay and the related monitoring and modeling efforts; and
- Section IV.D provides an overview of U.S. estuary programs and some major efforts to characterize loadings of nitrogen and toxic pollutants to coastal waters.

Although this chapter describes current data and programs specific to the subject waterbody, much of the information is relevant to other waters as well. For example, those interested in smaller estuaries will gain insight from information presented in the Chesapeake Bay section.

The Great Lakes and Lake Champlain represent two important freshwater systems in the United States. Lakes are sensitive to pollution inputs because they lack any dominant, unidirectional flow, and as a result, there is a slow change of water and a resulting retention of pollutants.

The Great Lakes contain approximately one-fifth of the world's supply of fresh surface water. These lakes have played a vital role in the history and development of the United States and Canada. They are stressed by a wide range of pollution sources associated with the large urban centers located on their shores. Because the Great Lakes system is a relatively closed water system (very large volume, with relatively small water inflows and outflows), many of the pollutants that reach the Great Lakes remain in the system for extended periods of time. For example, Lake Superior replaces all the water in the lake every 191 years, Lake Erie every 2.6 years.

Lake Champlain is located in the northeastern United States, shared by the states of New York and Vermont and the Province of Quebec. Although much smaller in surface area than the Great Lakes, Lake Champlain is still one of the largest freshwater lakes in the United States and its natural resources are important to the local economy. The Lake Champlain basin, or watershed, is much larger relative to its water surface area than the Great Lakes, and so watershed throughput is much more of an issue for Lake Champlain. Toxic pollutants are an issue of wide public concern in the Lake Champlain Basin, due in large part to fish consumption advisories for PCBs and mercury issued by both New York and Vermont, and the potential impact on drinking water and the Lake's many other uses.

"Coastal waters," for the purposes of CAA section 112(m), are defined as those estuaries designated for the National Estuary Program (pursuant to section 320(a)(2)(A) of the federal Water Pollution Control Act) or designated for the National Estuarine Research Reserve System (pursuant to section 315 of the Coastal Zone Management Act). Chesapeake Bay is identified by name in section 112(m).

Estuaries occur where rivers empty into the ocean, mixing together fresh water and salt water, and creating an ecosystem distinct from, and often more productive than, either fresh or salt water systems. Estuarine waters include bays, sounds, marshes, swamps, inlets, and sloughs. These environments are characterized by varying degrees of salinity, high turbidity levels, and complex water movement affected by ocean tides, river currents, and wind. Estuaries are critical coastal habitats that serve as spawning grounds, nurseries, shelters, and food sources for many different species of shellfish, fish, birds, and other wildlife. The leading environmental problems in estuarine systems at present are eutrophication,[10] contamination by toxic chemicals and pathogens (disease-causing organisms), over-harvesting, and loss of habitat.

In 1975, Chesapeake Bay became the nation's first estuary to be targeted for protection and restoration. Over the past decade, other coastal programs, such as the National Estuary Program, the National Estuarine Research Reserve System, and the Gulf of Mexico Program, have been established to protect and restore water quality and living resources in U.S. estuaries and coastal waters. Chesapeake Bay was also among the first estuaries where atmospheric sources of nutrients and toxic pollutants were recognized as significant inputs to the waterbody. Recently, research on other U.S. coastal waters has begun to evaluate the loadings of nutrients and toxic pollutants to their watersheds from atmospheric sources. The Great Waters program has focused primarily on Chesapeake Bay for estuarine issues, and has found that information developed for this waterbody is generally applicable to several other East Coast estuaries, when

[10] As discussed in Section II.D and in this chapter, eutrophication is over-enrichment of waters that is characterized by algae blooms, turbid waters, and low or no dissolved oxygen conditions.

adjustments are made for respective waterbody's physical, chemical, and geomorphological characteristics.

IV.A The Great Lakes

The Great Lakes, comprised of Lakes Superior, Michigan, Huron, Erie, and Ontario, are an important part of the physical, cultural, and industrial heritage of North America (see Figure IV-1). The Great Lakes ecosystem, the interacting components of air, land, water, and living organisms, including humans, is one of the largest surface systems of freshwater on earth. This ecosystem contains 18 percent of the world's freshwater supply and 95 percent of the surface freshwater within the United States. Only the polar ice caps and Lake Baikal in Siberia contain more freshwater than the Great Lakes. By virtue of their size, the Great Lakes affect the climate of the surrounding region. Areas of Michigan, Ontario, and New York generally have warmer, though snowier, winters than other parts of North America at similar latitudes because, as a result of little continual current, the lakes retain a large amount of heat. In spring and early summer, the lakes are slow to warm, thereby keeping the nearby land areas cool.

The Great Lakes sustain a rich diversity of fish, birds, and other wildlife. Native fishes important for commercial and recreational harvest include lake trout, lake whitefish, and walleye. Non-native species such as smelt, white perch, brown trout, rainbow trout, and several Pacific salmon species also contribute substantially to the total annual fish harvest. Approximately three million waterfowl follow the Atlantic and Mississippi flyways through the Great Lakes basin each year. Native animals include deer, fox, moose, wolves, beaver, mink, and muskrat. In addition, the Great Lakes ecosystem supports more than 100 globally endangered or rare species (Nature Conservancy 1994).

Economic Highlights of the Great Lakes

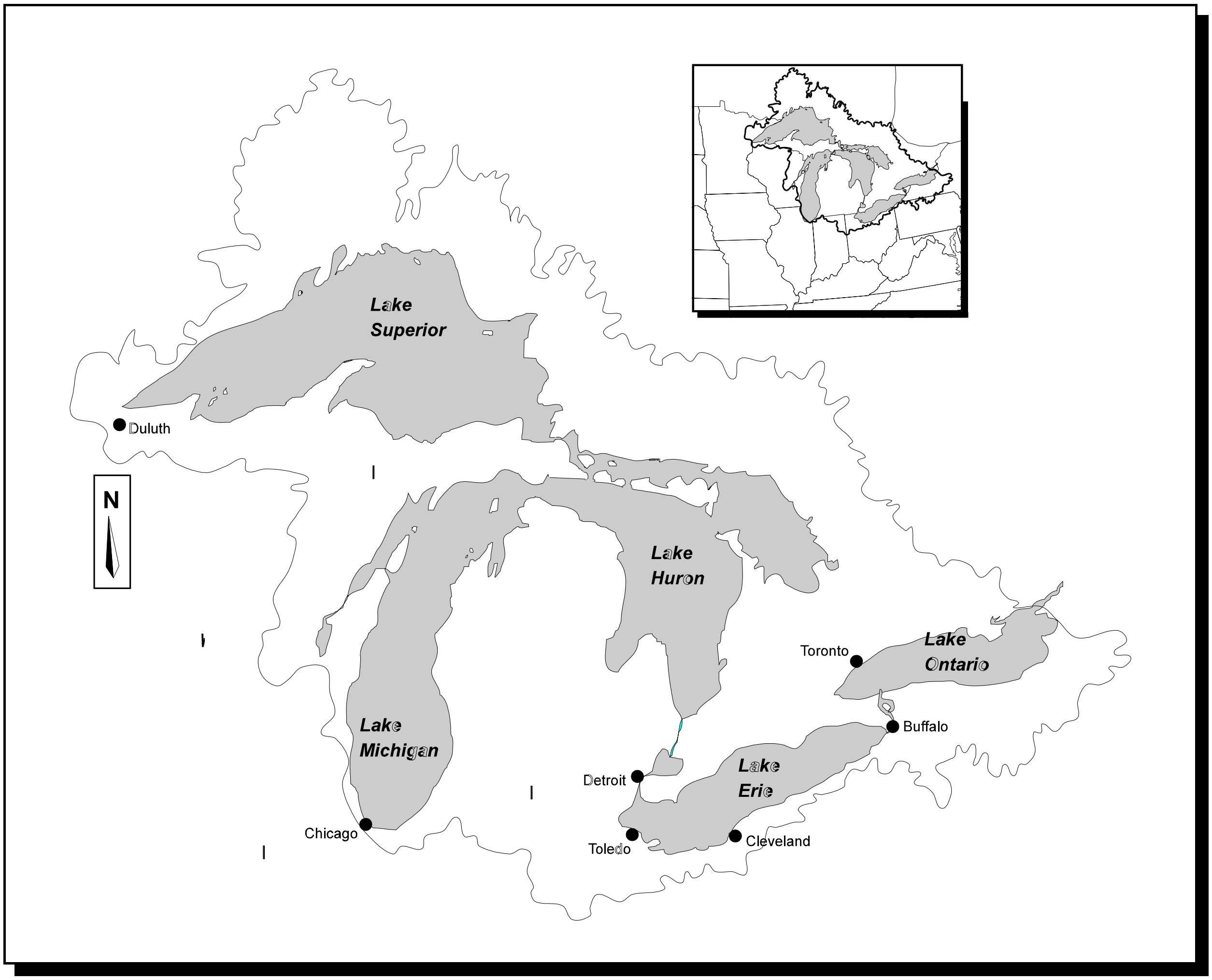
- Approximately 11% of total employment and 15% of manufacturing employment of combined U.S. and Canadian workers are located in the Great Lakes basin.
- Trade between Canada and the eight Great Lakes States in 1992 was valued at $106 billion (56.2% of the U.S.-Canada total).
- An estimated 900,000 to 1 million U.S. and Canadian boats operate each year, resulting in a direct spending impact on the regional economy of more than $2 billion.
- About 2.55 million U.S. anglers fish the Great Lakes; total trip-related and equipment expenditures were $1.33 billion in 1991.

Source: Allardice and Thorp 1995.

The Great Lakes basin is home to more than 33 million people, including 10 percent of the U.S. population and 25 percent of the Canadian population. Over 23 million of these people depend on the Great Lakes for drinking water. Industries use the water to make products, to cool manufacturing processes or power generation equipment, and to ship raw materials and finished products. Residents and visitors alike enjoy an abundance of recreational activities, including boating, swimming, fishing, sightseeing, camping, and hiking.

The concentration of human activities in the Great Lakes basin (e.g., manufacturing, transportation, agriculture, fishing) imposes stresses on the ecosystem and has prompted significant concerns for the health and well-being of the human residents. Many of the major stressors and resultant effects were documented in the First Great Waters Report to Congress. The current report builds on this information and presents some of the potential problems that may affect the Great Lakes basin from the perspective of the CAA, and major programs that are underway to address those problems.

FIGURE IV-1
Great Lakes Basin



Physical Features of the Great Lakes						
	Superior	**Michigan**	**Huron**	**Erie**	**Ontario**	**Totals**
Volume (km^3)	12,100	4,920	3,540	484	1,640	22,684
Maximum Depth (meters)	406	282	229	64	244	NA
Water Area (km^2)	82,100	57,800	59,600	25,700	18,960	244,160
Land Drainage Area (km^2)	127,700	118,000	134,100	78,000	64,030	521,830
Retention Time (years)	191	99	22	2.6	6	NA

NA = not applicable

The remainder of Section IV.A presents:

- Current knowledge and recent measurements of atmospheric levels and deposition of toxic pollutants to the Great Lakes;

- Information on major activities/programs currently in progress to assess atmospheric deposition of air pollutants to the Great Lakes;

- Efforts supported by the United States, as well as Canada, to reduce and mitigate atmospheric emissions in the Great Lakes basin; and

- Brief discussion on current information gaps, and future research needs to improve understanding of atmospheric deposition of pollutants into the Great Lakes.

Common Terminology for Pollutant Movement in a Waterbody

FLUX

Transport of a chemical across an interface (e.g., between air and water) for a given area and time, accounting for both inputs and outputs. Net flux is equal to all positive loadings minus all negative loadings.

INPUTS (positive loading)

Wet Deposition: Gases and particles carried in precipitation (rain, snow, sleet) and deposited on land and water surfaces.
Dry Particle Deposition: Pollutants, bound to particles, deposited on land and water surfaces in the absence of precipitation.
Gas Absorption: Gaseous form of pollutants crossing air-water interface into the water (portrayed as a positive number in a flux calculation).
Waterborne Discharge: Pollutants discharged directly to water (e.g., by industrial discharge, urban storm-runoff).
Tributary Loading: Pollutants entering waterbody through connecting channels, streams, and rivers.

OUTPUTS (negative loading)

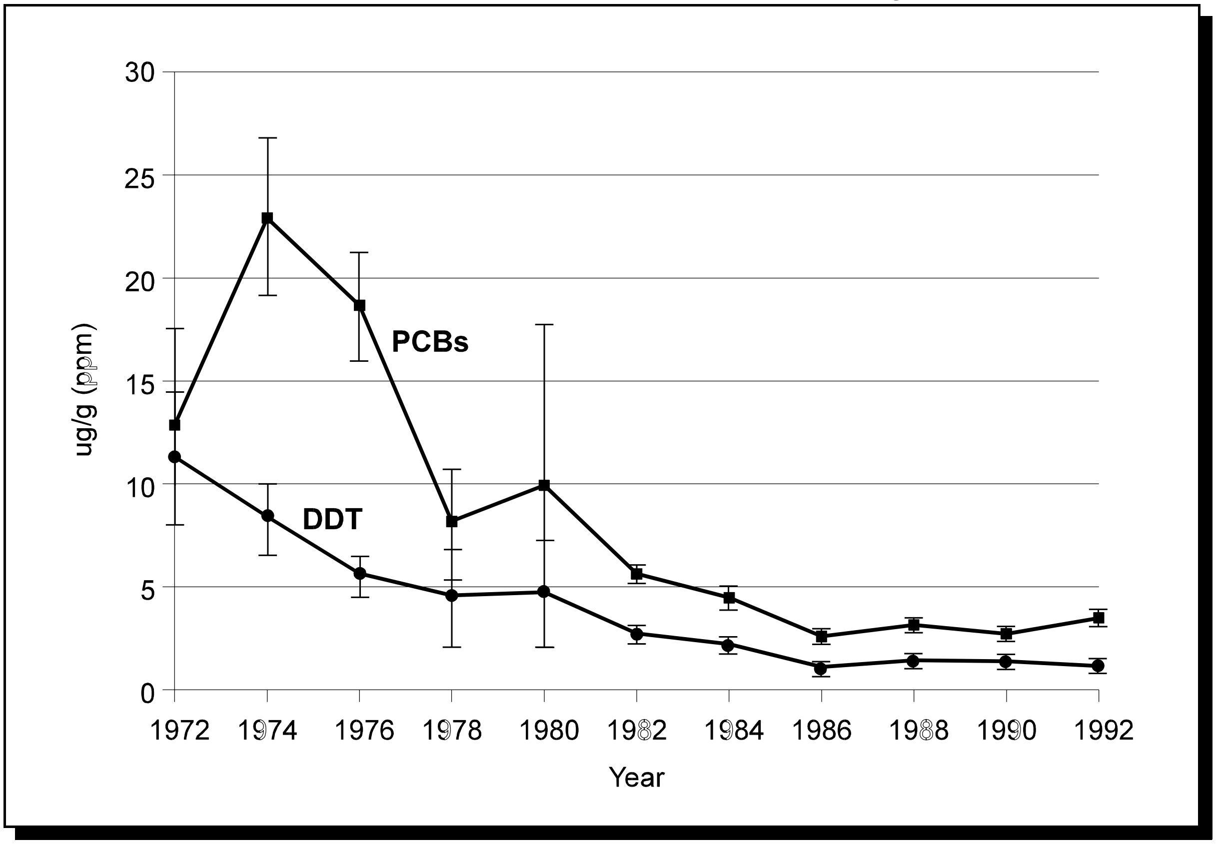
Volatilization or Gas Evasion: Gaseous form of pollutants crossing air-water interface into the air (portrayed as a negative number in a flux calculation).
Sedimentation: Settling of particles by gravity to bottom sediments.
Outflow: Pollutants flowing with water out to rivers or to the ocean.

Atmospheric Deposition of Great Lakes Contaminants

Hundreds of anthropogenic chemicals have been identified in the Great Lakes ecosystem. High levels of certain bioaccumulative pollutants remain in certain fish and wildlife species, and fish advisories have been issued by many Great Lakes states for several pollutants of concern, specifically chlordane, dioxins, mercury, PCBs, and toxaphene (specific advisories are listed in Appendix B). For example, although concentrations of PCBs and DDT in Lake Michigan lake trout are currently about one-tenth of those of 20 years ago (Figure IV-2), the concentrations are still at levels that warrant issuance of public health advisories regarding the consumption of these fish. Advisories may especially apply to specific subpopulations, such as children and women who are pregnant or anticipate bearing children.

The pollutants of concern have been associated with health problems in certain fish and wildlife species, although with the decline of some pollutant levels, many species may be recovering. For example, the number of double-crested cormorants living on the Great Lakes has increased more than 20-fold during the past 15 years. Prior to this, numbers of these fish-eating birds declined during the 1970s due to reproductive failure from DDE-induced egg shell thinning. Health problems persist for fish and wildlife in certain locations, particularly in waters

Figure IV-2
PCBs and DDT in Lake Trout from Lake Michigan



with highly contaminated bottom sediments, and for predators high in the food web, such as lake trout, mink, and bald eagles.

During the 1980s, studies in the Great Lakes showed that atmospheric deposition may be a major route of introduction of a number of pollutants to the Great Lakes. For example, atmospheric transport of toxaphene was implicated when the insecticide was found in fish in Lake Siskiwit, located on an island in Lake Superior Because the elevation of Lake Siskiwit is above that of Lake Superior, it does not receive any groundwater from Lake Superior and thus pollutant input. This pesticide was used mainly on cotton crops in the southern United States, prior to its cancellation in 1982 (McVeety and Hites 1988).

As a result of this and other findings, the United States and Canada established a joint monitoring network called the Integrated Atmospheric Deposition Network (IADN). The IADN is designed to assess the magnitude and trends of atmospheric deposition of target chemicals to the Great Lakes, and to determine emission sources wherever possible. The program responds to the Great Lakes Water Quality Agreement (GLWQA) between the United States and Canada, specifically to the needs of Annex 15 which addresses issues concerning airborne contaminants in the Great Lakes basin. A more detailed discussion of the rationale, design, and results of the IADN, as well as the uncertainties that exist in calculating atmospheric deposition estimates, is presented in the following subsection (Program Actions to Characterize Atmospheric Contamination in Great Lakes).

TABLE IV-1
Atmospheric Loading Estimates for Selected Pollutants (kg/year) in the Great Lakes

Pollutant of Concern	Superior	Michigan	Huron	Erie	Ontario
PCBs[a] (Wet and Dry)					
1988	550	400	400	180	140
1992	160	110	110	53	42
1994	85	69	180	37	64
PCBs[a] (Net Gas Transfer)[b]					
1988	-1900	-5140	-2560	-1100	-708
1994	-1700	-2700	--	-420	-440
DDT (Wet and Dry)					
1988	90	64	65	33	26
1992	34	25	25	12	10
1994	17	32	37	46	16
DDT (Net Gas Transfer)					
1988	-681	-480	-495	-213	-162
1994	30	67	--	34	13
Benzo(a)pyrene (Wet and Dry)					
1988	69	180	180	81	62
1992[c]	120	84	84	39	31
1994	200	250	--	240	120

-- Not determined or reported.
[a] Data presented for PCB congeners 18, 44, 52, and 101 (each with 3-5 chlorines in chemical structure).
[b] The convention is to assign a negative number to loss of pollutant from the lake (i.e., volatilization). Thus, the resulting number expresses the mass of a pollutant going into or coming out of the lake per year (i.e., a positive net gas transfer indicates a net input of the pollutant to the lake and a negative net gas transfer indicates a net loss or output from the lake).
[c] Data from 1992 may represent an underestimation in the measurement of benzo(a)pyrene.

Sources: Eisenreich and Strachan 1992; Hillery et al. 1996; Hoff et al. 1996; and Strachan and Eisenreich 1988.

The first consensus data report for IADN (Eisenreich and Strachan 1992) revised and improved the very broad estimates of atmospheric deposition of toxic contaminants that were previously compiled (Strachan and Eisenreich 1988). More recent data were incorporated into deposition estimates for 1994 and compared to the earlier results (Hoff et al. 1996), as shown in Table IV-1. A general decline in some pollutant levels is suggested from the estimates in Table IV-1. Average estimated atmospheric loadings of certain pollutants to the five Great Lakes between 1991 and 1993 are presented in Table IV-2. The data in Tables IV-1 and IV-2 may not be comparable because the estimates represent measurements at different time frames. It is important to recognize that the values presented in these tables are based on preliminary data and limited samples. Furthermore, contributions from water inputs and outputs are not included. As such, overall loadings to the lakes cannot be established from these data alone.

Atmospheric loadings of pollutants are calculated using atmospheric concentration data gathered by IADN and estimates for various parameters such as lake surface area. The wet deposition data are based on estimated annual precipitation rates, and do not use actual rainfall amounts. Although uncertainties exist for the parameters which can lead to some degree of error, the atmospheric deposition estimates are based on the best scientific data currently available.

The remainder of this subsection presents atmospheric concentration and deposition data collected primarily from IADN on some pollutants of concern, as well as information on current trends of pollutant deposition. The pollutants of concern discussed include PAHs, PCBs, pesticides (e.g., DDE, DDT, lindane, toxaphene), and trace metals (e.g., lead, mercury). PCBs,

TABLE IV-2
Average Estimated Atmospheric Loadings[a] of Selected Pollutants to the Great Lakes (kg/year) (1991-1993)

Atmospheric Process[b]	Lake Superior	Lake Michigan	Lake Huron	Lake Erie	Lake Ontario
PCBs					
Wet deposition	58	52	180	21	58
Dry deposition	27	16	---	16	5.7
Net gas transfer[c]	-1700	-2700	---	-420	-440
Absorption	320	390	---	340	130
Volatilization	-2000	-3100	---	-760	-560
Dieldrin					
Wet deposition	21	58	10	28	11
Dry deposition	7.4	8	---	5.6	1.7
Net gas transfer	-780	---	---	-610	-320
Absorption	120	200	---	67	43
Volatilization	-910	---	---	-680	-370
DDE					
Wet deposition	2.6	3.8	10	4.6	4.5
Dry deposition	0.4	0.5	---	0.5	0.2
Net gas transfer	---	---	---	---	-170
Absorption	10	26	---	14	12
Volatilization	---	---	---	---	-180
Lindane					
Wet deposition	62	65	140[d]	46	51
Dry deposition	0.6	1.1	---	0.4	0.2
Net gas transfer	140	1200	---	61	16
Absorption	340	1400	---	180	72
Volatilization	-200	-140	---	-110	-56
Benzo(a)pyrene					
Wet deposition	140	170	---	180	56
Dry deposition	58	77	---	63	60
Net gas transfer	87	---	---	---	---
Absorption	100	92	---	51	7.5
Volatilization	-17	---	---	---	---

[a] Values calculated from atmospheric loading equations, and based on atmospheric concentration data collected from Integrated Atmospheric Deposition Network (IADN); summarized from Hoff et al. (1996).
[b] Wet deposition based on estimated annual precipitation rates, and does not use actual rainfall amounts. Dry deposition represents only data for particle form of pollutant (i.e., gaseous form included in absorption values).
[c] Net gas transfer is the sum of gas absorption and volatilization. Water concentration data are taken from past literature and compared with the more recent air measurements, which may lead to some potential error in gas transfer estimates. Values for net gas transfer are rounded off and thus estimates may not add up in the table.
[d] High estimated value may be due to very limited number of samples for 1992 season and should be reconsidered as more data become available (Hoff et al. 1996).
--- Not determined or reported.

toxaphene, and mercury are given greater focus because fish advisories are currently issued for these pollutants. Fish advisories also exist for chlordane and dioxins for the Great Lakes, but they are not addressed in this section because recent information is limited. Potential sources of the pollutants are also discussed, as well as uncertainties in the data.

PAHS

Polycyclic aromatic hydrocarbons (PAHs), a subset of POM, are a class of semi-volatile compounds produced in combustion processes and are widely distributed in the environment. As indicated in the First Report to Congress, approximately 72 to 96 percent of the total annual loading of one common PAH, benzo(a)pyrene, to Lakes Superior, Michigan, and Huron is attributed to atmospheric deposition.

PAHs are detected both in the gaseous and particulate phases, but some of the most toxic PAHs are largely in the particulate phase in the atmosphere. For the most toxic PAHs, dry deposition is generally the main route of deposition to the lakes (Hoff and Brice 1994). For benzo(a)pyrene, however, wet deposition seems to be the major source of atmospheric loadings to Lake Michigan in all seasons of the year. The IADN data suggest that, for Lake Superior, the net movement of the gaseous phase benzo(a)pyrene is largely to the water; data are limited for the other lakes (see Table IV-2). Comparing recent wet and dry deposition values with historic data, the loading of benzo(a)pyrene to the lakes appears to have increased (Table IV-1). However, the 1992 finding may be attributed to an underestimation in the measurement of benzo(a)pyrene (Hoff et al. 1996).

A recent study found that total wet and dry deposition for benzo(a)pyrene was 50 times higher at an urban site (Chicago) than at remote IADN sites in Lakes Michigan and Superior (Sweet and Harlin 1996). The investigators concluded that large areas of Lake Erie, Lake Ontario, and southern Lake Michigan have elevated PAH deposition rates due to emissions from nearby urban areas. Although the total deposition of PAHs are lower in rural than urban sites, the relative amounts of the individual PAHs (i.e., relative ratios of the individual PAHs) is very similar at urban and nonurban sites, suggesting that little chemical degradation occurs during transport of PAHs from urban source areas to rural and remote sites several hundred kilometers away.

PCBS

PCBs are a class of highly toxic, persistent, and bioaccumulative chemical compounds. PCBs in Great Lakes fish have long been linked to developmental and growth problems in infants born to women who regularly consumed PCB-contaminated fish in the late 1970s. PCBs were produced from 1927 to 1977 for the purpose of insulating and cooling electrical equipment. In the late 1970s, Monsanto Company, sole manufacturer of PCBs in the United States, voluntarily stopped production of PCBs. Estimates suggest that 282 million pounds of pure PCBs -- 20 percent of PCBs ever produced -- were still in service at the end of 1988.

PCBs manufactured before production was stopped are still found in the Great Lakes. They are present in older commercial and industrial equipment (e.g., transformers, capacitors). There are no phaseout deadlines that require removal of the equipment to avoid breakage and release, although this equipment is tightly regulated under the Toxic Substances Control Act (TSCA). As a result of past use and disposal practices, PCBs may reside in sediments in surface waters and in other areas, such as waste sites. As the contaminated sediment is disturbed, the

PCBs may be re-released and resuspended in the water, allowing for continued bioaccumulation in Great Lakes fish. Remediation programs are in-place to address PCB-contaminated waste sites. Other continuing PCB sources include unregulated sources that potentially contain PCBs and releases, as well as releases by those PCB owners who are not aware of the presence of PCBs or of the special management requirements for PCB-containing equipment.

Despite the fact that PCBs are one of the most tightly regulated and controlled group of pollutants under federal regulatory programs, fish consumption advisories still exist for PCBs in all five of the Great Lakes (see Appendix B). For example, although PCB levels have declined in Lake Michigan water, there has been a constant or increasing level of PCBs in some Lake Michigan fish in the last few years, possibly due to resuspension from sediment or from changes in the Lake Michigan food chain (see Section II.B).

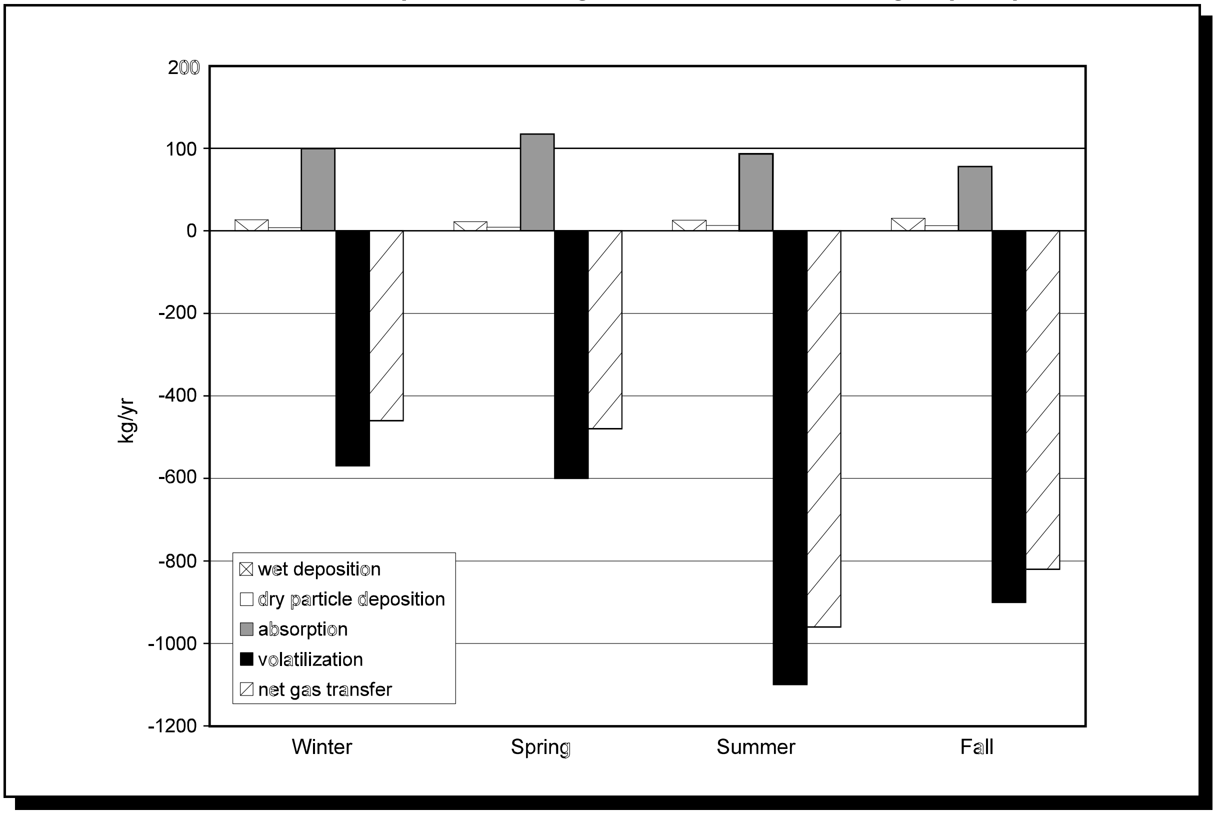
Volatilization is the dominant mechanism in air-water gas exchange of PCBs (Table IV-2). Volatilization of PCBs from the Great Lakes is estimated to be as high as 3,100 kg/year for Lake Michigan and 2,000 kg/year for Lake Superior. In contrast, estimates of wet deposition of PCBs are less than those for volatilization and are nearly the same for Lakes Superior, Michigan, and Ontario (52-58 kg/year) with Lake Erie showing a lower rate (21 kg/year) and Lake Huron showing the highest rate (180 kg/year) (Table IV-2). Dry deposition rates of PCBs are similar in Lakes Superior, Michigan, and Erie (16-27 kg/year), with Lake Ontario showing a lower rate (5.7 kg/year). As presented in Table IV-1, from 1988 to 1994, wet and dry deposition of PCBs to each of the Great Lakes has decreased. Therefore, the net loss to the atmosphere would suggest that the amount of PCBs in water is declining. Tables IV-1 and IV-2, however, represent data only for the atmospheric movement of pollutants and therefore, do not indicate the waterborne inputs to each lake (such as particles in the water, industrial water discharges, and especially urban storm-runoff which goes directly into the lakes). Also, this study on gas exchange is preliminary and there are considerable uncertainties in the estimates. Additional work addressing the uncertainties and other routes of pollutant movement may give a different balance, although the importance of gas exchange is clear.

Wet and dry deposition of PCBs are similar over seasons, while net gas exchange is highly seasonal, exhibiting much greater effect with high temperatures (Figure IV-3). To date, no studies on seasonal variation in PCB concentrations in water have been published to compare with the seasonal atmospheric loading data. As stated earlier, uncertainties exist in calculating deposition estimates, since some estimates are based on rough approximations or assumptions using the best science available at this time.

PESTICIDES

Volatilization of the pesticides dieldrin and DDE (a metabolite of DDT) in many of the Great Lakes is a significant process. Net atmospheric loading is negative, indicating that movement of these two pesticides between air and water is mostly volatilization (Table IV-2). Fluctuations in gas equilibrium conditions may be influenced by the water concentration data, differences in temperature, and/or errors in the Henry's Law constant used in calculating gas movement. For example, DDT net gas transfer estimates are hindered by difficulties in obtaining precise water concentration data because DDT levels in the lakes are close to the analytical detection limit. From Table IV-1, DDT wet and dry deposition loadings declined between 1988 and 1992, but rose slightly for all lakes except Lake Superior in 1994 (Hillery et al. 1996).

FIGURE IV-3
Seasonal Atmospheric Loadings of PCBs in Lake Michigan (1994)



Wet and dry deposition of lindane appeared to be fairly uniform across all lakes (see Table IV-2). Gaseous lindane generally seemed to be in equilibrium within Lakes Erie and Ontario, while gas absorption is the dominant mechanism in air-water exchange for Lakes Superior and Michigan (Table IV-2). The net gas transfer of lindane in Lake Michigan is into the lake in the winter and spring and out of the lake in the summer and fall. For many pesticides, gas transfer is strongly dependent on seasons, with net outputs in the summer and net inputs in the winter (Achman et al. 1992; Hoff et al. 1993; McConnell et al. 1992; Ridal et al. 1996).

Toxaphene in the Great Lakes Basin. Toxaphene, a semi-volatile insecticide containing a mixture of chlorinated bornanes (class of aromatic hydrocarbons), has been recognized as one of the contaminants with the highest concentrations in Great Lakes fish (Ribick et al. 1982; Schmitt et al. 1981, 1985, 1990). Because of its volatility and persistence, toxaphene is still widely distributed through the atmosphere, even though it is no longer used in the United States (Rapaport and Eisenreich 1986). Toxaphene was been found to be a major contaminant in lake trout and whitefish from Siskiwit Lake on Isle Royale, Lake Superior (De Vault et al. 1996) (see sidebar).

Toxaphene in Lake Trout

Since 1991, the state of Michigan has issued a consumption advisory for Siskiwit lake trout from Lake Superior based on exceedance of the FDA's 5.0 ppm action level for toxaphene. In 1995, the Canadian Province of Ontario issued fish consumption advisories for several different species in Lake Superior and upper Lake Huron, triggered by their toxaphene levels and a lowering of Health Canada's action level for toxaphene to 0.2 ppm.

Toxaphene's discovery on pristine Isle Royale, exposed only to atmospheric deposition, seemed indicative of long-range transport via the atmosphere since it had been used primarily as a pesticide in the southern United States (Hoff et al. 1993). This hypothesis has been supported by studies that found toxaphene concentrations in Canadian air masses that had originated in the southern United States. However, there is also evidence to suggest that some of the toxaphene found in Lake Superior and northern Lake Michigan may have local origins. A study that analyzed fish from rivers in the southeastern United States, the Great Lakes, and Isle Royale, collected during 1982, found differences in composition of toxaphene in fish between sites, suggesting that potential local influence may be important, rather than long-distance atmospheric transport from the southeastern United States to the Great Lakes (Petty et al. 1987).

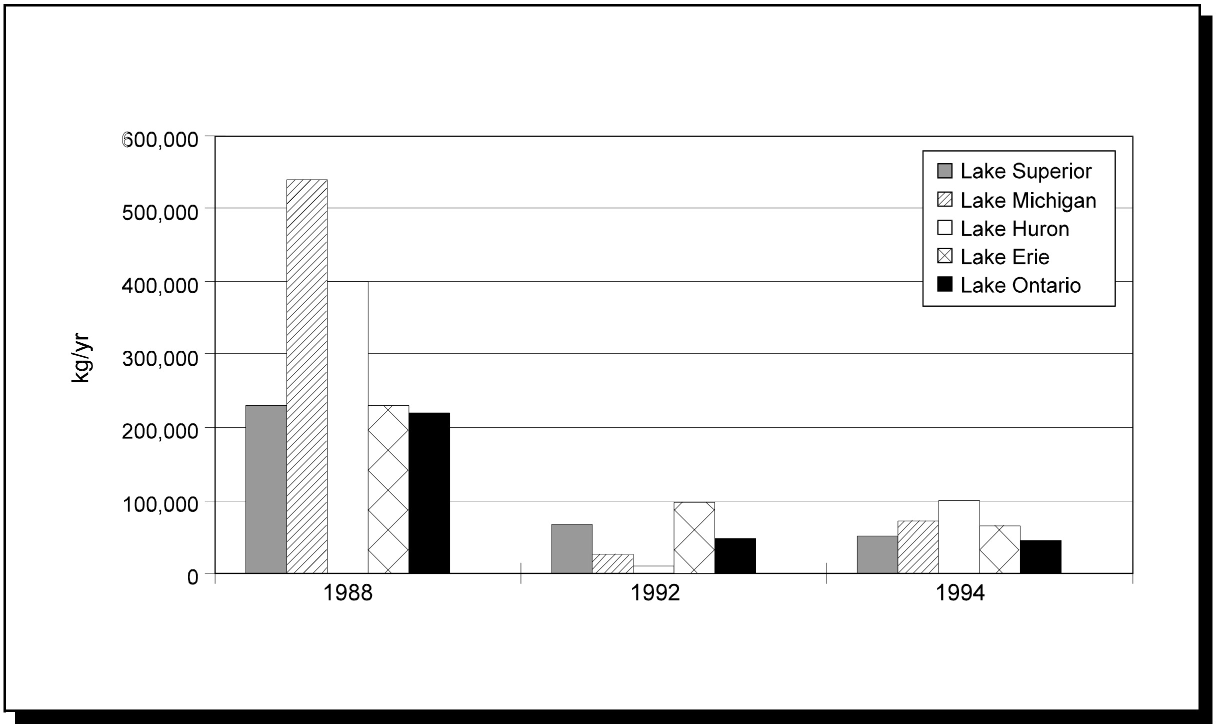
EPA recently supported monitoring of toxaphene in Great Lakes fish and sediment which has revealed two trends. First, there has been a statistically significant decline in the concentration of toxaphene in fish from most waters, as might be expected following reduced use and later cancellation of the pesticide. Second, there has been no discernible decline in toxaphene levels in Lake Superior lake trout; toxaphene levels are higher than levels of other measured contaminants in fish from anywhere in the Great Lakes.

At this time, there are several hypotheses for the relatively elevated levels of toxaphene observed in Lake Superior and northern Lake Michigan. First, the continued use of toxaphene by other countries and subsequent atmospheric transport to the Great Lakes basin may increase levels. Another possibility is the previous local use of the pesticide. Toxaphene was once used to kill undesirable fish communities (Lockhart et al. 1992; Stern et al. 1993). This practice occurred in parts of Canada and the northern United States for fish restocking on small glacial lakes; it was applied to at least 80 lakes during the 1950s and 1960s in Wisconsin (Hughes 1968). It has also been proposed that Lake Superior lake trout may be slower to reflect a decrease in contaminant levels in their food web due to their greater age; however, Glassmeyer et al. (1997) found that toxaphene levels in Lake Superior fish were still elevated compared to levels in fish from the other lakes. Another explanation that has been suggested is that toxaphene persists longer in colder, less productive waters such as Lake Superior. Finally, the high toxaphene levels may be a result of the release of toxaphene into the waters as a byproduct in the production of paper; there are 74 pulp and paper mills that directly discharge to all the Great Lakes (IJC 1995), with the paper industry most concentrated near Lake Superior and upper Lake Michigan (Green Bay).

TRACE METALS

A number of trace metals are of concern in the Great Lakes, though new data are limited. Data from 1994 suggest that wet deposition is the dominant atmospheric transport mechanism for trace metals to the Great Lakes (Hoff and Brice 1994). The most consistent trend in the deposition of trace metals was the reduction in lead in 1994 compared with 1988 values for all the lakes (Figure IV-4). This finding is not surprising given the phaseout of leaded gasoline in the United States beginning in the 1970s and accelerating in the mid-1980s. The gaseous phase of lead is assumed now to be negligible. Arsenic deposition also has decreased. The reason for this finding is not as clear but it has been hypothesized that process changes by Noranda, a major emitter of arsenic in Canada through mining, smelting, and refining of metal products, may have led to the decline.

FIGURE IV-4
Atmospheric Loadings of Lead to the Great Lakes (1988-1994)



Mercury in the Great Lakes Basin. Currently, six of the eight Great Lakes states (Michigan, Ohio, Wisconsin, New York, Pennsylvania, and Minnesota) have issued advisories restricting consumption of fish from some state waters due to mercury contamination. Mercury contamination, or high mercury levels in fish tissue, is also the most frequent basis for fish advisories issued by the Province of Ontario. Many of their advisories are applicable to areas of the Great Lakes.

During the early 1970s, mercury was found in fish from Lake Huron, Lake St. Clair, western Lake Erie, eastern Lake Ontario, and the St. Lawrence River at levels that led the United States and Canada to close commercial fisheries. Subsequently, mercury levels fell in these waters, because of modification or closure of certain chloralkali facilities and pulp and paper mills whose wastewater discharges contained large quantities of mercury. In 1970, mercury levels in Lake St. Clair walleye were 2 parts per million (ppm); by the mid-1980s, levels in these walleye had subsided to 0.5 ppm (Environment Canada et al. 1991). There are other signs of reduced mercury levels in the Great Lakes through dated sediment cores and populations of smelt in the lakes.

Because of large direct discharges of mercury being terminated following implementation of the Clean Water Act (CWA), the atmosphere is now the dominant pathway by which mercury reaches the Great Lakes. Currently, the best estimate of atmospheric deposition to the five Great Lakes is approximately 6,800 kg per year (15,000 pounds) (Eisenreich and Strachan 1992). Loadings of mercury to Lakes Superior and Michigan are primarily from the atmosphere. For Lake Ontario, the percentage of atmospheric contribution of mercury is relatively modest because the lake receives mercury from waters that flow from the upper lakes (Sitarz et al. 1993).

Even some of the mercury borne to the Great Lakes via their tributaries includes contamination previously deposited from the atmosphere to their watersheds.

Unlike other trace metals, mercury exists in the air predominantly in the gaseous phase due to its volatility. Estimates of wet and dry deposition of mercury to Lake Superior are about five times higher than net gas transfer to the atmosphere. The net annual atmospheric loading of mercury to Lake Superior is calculated to be about 635 kg/year (Hoff et al. 1996), which is comprised of the following estimates:

- 560 kg/year as wet deposition;
- 250 kg/year as dry deposition;
- 65 kg/year as absorption; and
- -240 kg/year as volatilization.

In an earlier study, gaseous phase mercury in the atmosphere was 1.57 ng/m^3, particulate phase mercury, 0.02 ng/m^3, and precipitation mercury, 10.5 ng/L, at a northern Wisconsin site near Lake Michigan (Fitzgerald et al. 1991). Comparison of these values with other U.S. sites is presented in Table IV-5 in Section IV.B.

In Michigan, atmospheric concentrations and wet deposition of mercury have been observed to vary geographically. Northern Michigan received only one-half the wet deposition of mercury deposited to southern portions of the state. Wet deposition varied by season, with mercury concentrations in precipitation two times greater during spring and summer than during winter. Higher levels of particulate mercury were observed in large urban areas. Modeling indicated that the dominant sources of mercury were located mostly to the south and west of Michigan (Keeler and Hoyer 1997).

Program Actions to Characterize Atmospheric Contamination in the Great Lakes

Research has occurred in the past few years to increase understanding of the effects, fate, and transport of toxic substances in the Great Lakes ecosystem. These efforts are designed to provide information to further characterize, as well as reduce, atmospheric contamination in the Great Lakes region. Some of the programs to assess the extent of atmospheric contamination in the Great Lakes basin are described below. At this time, many of these projects are collecting and/or compiling data, and results are not yet available for evaluation. Also, several notable programs/activities have been introduced in recent years to begin to reduce loadings and to mitigate existing contamination and are discussed in the following section (Toxics Reduction Efforts in the Great Lakes).

LAKE MICHIGAN MONITORING PROGRAM

A monitoring program for Lake Michigan has been implemented by EPA's Great Lakes National Program Office to support a number of activities that address reductions in the release of toxic substances, particularly persistent, bioaccumulative substances, to the Great Lakes system. The program is a key element of the Lakewide Management Plan (LaMP) for Lake Michigan (see next subsection, Toxics Reduction Efforts in the Great Lakes, for general information about the objectives of LaMPs).

The water quality criteria and values provided in the GLWQ Guidance, once adopted by the Great Lakes states, would apply to the entire Great Lakes system, regardless of the source of pollutants to those waters. In this manner, the proposed water quality criteria and the measured values provide the basis for integrating actions carried out under the range of environmental programs available to federal, state, and tribal agencies to protect and restore the Great Lakes ecosystem. The mass balance approach will facilitate this integration by evaluating multi-media load reduction actions required to ensure that Lake Michigan water quality meets the Great Lakes water quality criteria (GLWQC).

The primary goal of the Lake Michigan Monitoring Program is to develop a sound, scientific base of information to guide future toxic load reduction efforts at federal, state, tribal, and local levels. In particular, the following specific objectives are identified:

- Evaluate relative loading rates of critical pollutants by medium (atmospheric deposition, contaminated sediments, tributaries) to establish a baseline loading estimate to gauge future progress;

- Develop the predictive ability to determine the environmental benefits of specific load reduction scenarios for toxic substances and the time required to realize those benefits, including evaluation of benefits of existing environmental statutes and regulations; and

- Improve our understanding of the key environmental processes that govern the cycling and bioavailability of contaminants within relatively closed ecosystems.

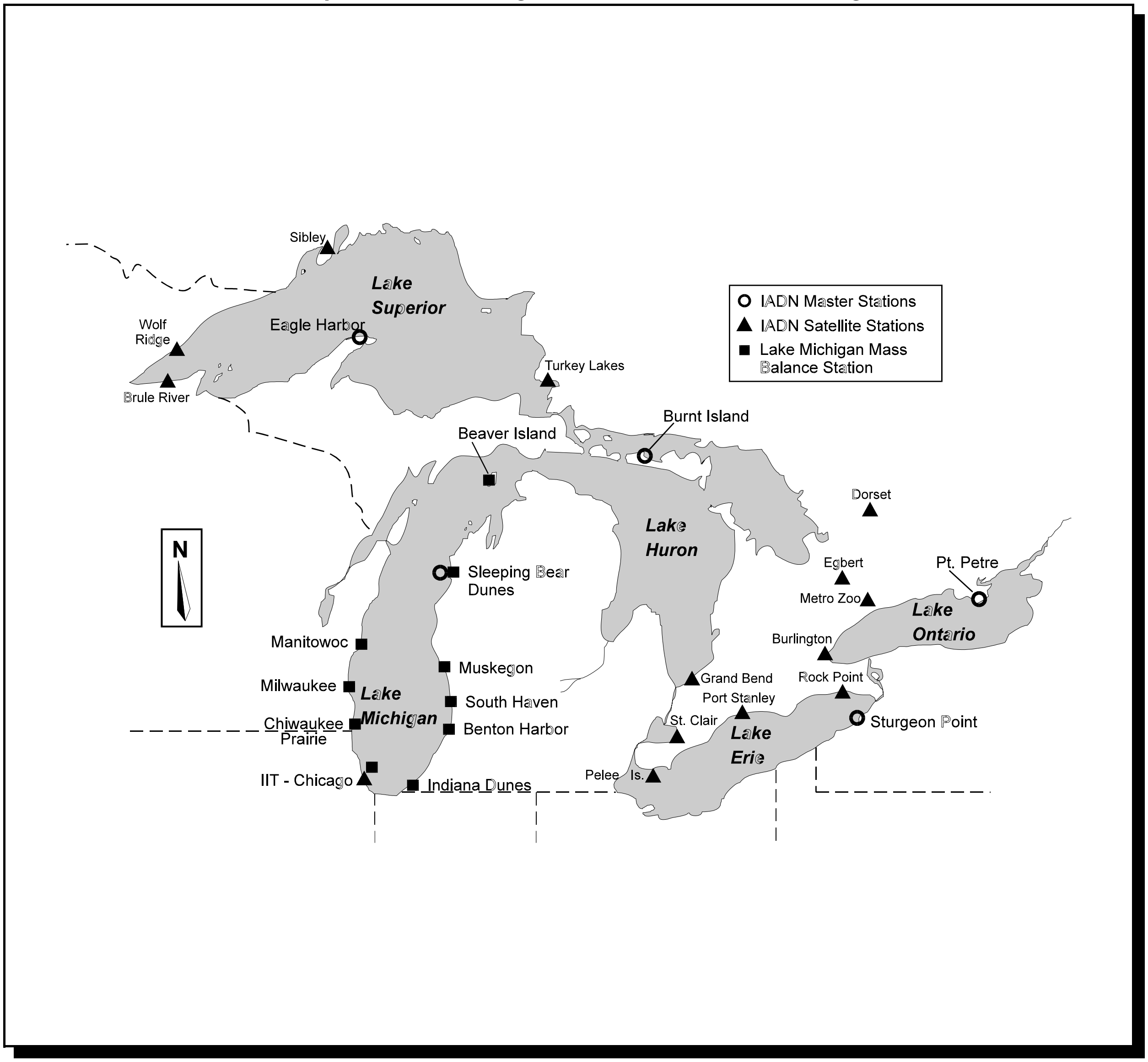
Lake Michigan Mass Balance Study. One of the ways to address the objectives of the Lake Michigan Monitoring Program, as well as to assist EPA in implementing section 112(m) of the CAA, is through a mass balance study. The mass balance study will characterize the loadings, transport, and fate of selected pollutants in a defined ecosystem, through monitoring and modeling. These measuring and estimating techniques can be applied to other ecosystems. EPA initiated the Lake Michigan Mass Balance Study, a comprehensive sampling effort that includes measurements of contaminants in the atmosphere, tributaries, lakewater, sediments, and food chain, to support model components. The atmospheric sampling sites for this mass balance study are shown in Figure IV-5.

Application of Modeling Tools from the Green Bay Mass Balance Study

In a pilot mass balance study by EPA and the Wisconsin Department of Natural Resources, water-insoluble organic compounds were monitored in Green Bay, Wisconsin, from 1988 to 1992. The analytical and modeling tools used in the study may be applied to the Great Lakes, Lake Champlain, and coastal estuaries. The **Lake Michigan Mass Balance study** is the first full-scale application of this methodology for toxic pollutants and will serve as the basis of any future mass balance efforts for persistent, bioaccumulative chemicals. Data collected for this study are anticipated in 1997.

The Lake Michigan Mass Balance model is constructed for a limited group of pollutants (PCBs, trans-nonachlor [a bioaccumulative component of chlordane], and total mercury) present in Lake Michigan at concentrations that pose a risk to aquatic and terrestrial organisms (including humans) within the ecosystem, or that may accumulate to problematic concentrations

FIGURE IV-5
Atmospheric Monitoring Sites in the Great Lakes Region



in the future and that can serve as examples for other chemicals. In addition, atrazine (under consideration for addition as a Great Waters pollutant of concern), a commonly used herbicide in the Great Lakes basin and elsewhere in the United States, is also included in the model. This herbicide has been reported at elevated concentrations in Lake Erie tributaries, in the open waters of the Great Lakes, and in the atmosphere over the lakes. The inclusion of this chemical will provide a model for the more reactive, biodegradable compounds in current use. The model will be less comprehensive than that for PCBs and trans-nonachlor, because atrazine does not appreciably bioaccumulate and it will not be analyzed in the food chain.

The chemicals chosen cover a wide range of chemical and physical properties and are representative of other classes of compounds that could pose water quality problems. This approach will allow modeling of many other chemicals with limited data. Resource limitations, quality assurance requirements, and analytical and data handling limitations preclude intensive monitoring and model calibration for more than the above described target chemicals. While mass balance modeling will focus on the above chemicals, the determination of loadings and concentrations for additional contaminants and compounds useful for source apportionment and deposition modeling will be undertaken as part of the Lake Michigan Monitoring Program.

The Lake Michigan Mass Balance Study includes an atmospheric monitoring component to address research issues concerning urban and atmospheric deposition and exchange processes. The data will be used to calculate atmospheric loads to the ecosystem. Within this air transport component, special studies are being performed to determine whether emissions of hazardous air pollutants from the urban coastal regions (Chicago, IL, and Gary, IN) contribute significantly to atmospheric deposition to the adjacent waterbodies. The objectives of these special studies are to (1) measure wet and dry deposition fluxes of urban contaminants, (2) determine contributions of urban source categories to measured concentrations and deposition rates, and (3) assess the extent of air-water exchange of contaminants.

Previous studies indicated that urban emissions have a large impact on atmospheric concentrations of air toxics and on atmospheric deposition to the Great Lakes. Dry depositional flux of PCBs from Chicago was shown previously to be three orders of magnitude higher than that of non-urban areas (Holsen et al. 1991). In addition, the Lake Michigan Urban Air Toxics Study demonstrated that concentrations of several pollutants were significantly higher in Chicago urban areas than at less urbanized sites (Keeler 1994). An intensive study was recently conducted around Chicago to assess the impact of the urban area on atmospheric deposition and exchange with Lake Michigan, with three land-based monitoring sites around Chicago and one over-water site on a research vessel approximately five miles off the shore of Chicago. The three sampling periods that occurred between 1994 and 1995 were designed to provide information to track atmospheric plumes over and across the lake. Wet deposition, dry deposition, and lake water were analyzed for semi-volatile compounds (such as PCBs and PAHs) and trace metals (such as arsenic, mercury, and lead). All samples were taken on the same day to provide information on air-water exchange of contaminants. Results from this study are expected in 1997, with modeling results expected in 1998. This study is expected to contribute useful information on urban impact to Lake Michigan, as well as to address process-oriented research issues and provide data in support of source apportionment and trajectory modeling.

INTEGRATED ATMOSPHERIC DEPOSITION NETWORK (IADN)

As mentioned earlier in this chapter, IADN is a long-term, binational program between the United States and Canada to assess the magnitude and trends of atmospheric deposition of target chemicals to the Great Lakes and to determine emission sources wherever possible. The program addresses the mandate of the Great Waters program and the needs of Annex 15 of the GLWQA between Canada and the United States. It is designed to (1) provide the necessary standardized methods, monitoring data, and loadings estimates to assess the relative importance of atmospheric deposition compared to other inputs, (2) determine temporal trends and geographic variations in deposition, and (3) ultimately provide information on sources of these atmospheric pollutants. It is a combination of a surveillance/ monitoring network and a research program. Its goals are source attribution, process identification, and assessment of atmospheric impacts on environmental systems. At this time, annual and seasonal averages have been completed for four years of IADN operation. Data for selected pollutants were presented earlier in this section. Additional details may be found in Eisenreich and Strachan (1992), Gatz et al. (1994), and Hoff et al. (1996).

Compounds Measured in IADN

- ***Highest priority group:*** PCBs, lindane, PAHs, and lead. These pollutants were chosen for the first phase of IADN (1990-1992) to demonstrate the feasibility and accuracy of sampling and analytical methods.
- ***Second priority group:*** Chlorinated pesticides (such as HCB, DDT/DDE/DDD, trans-nonachlor, methoxychlor, mirex, dieldrin, aldrin) and trace metals (such as arsenic, selenium, cadmium, and mercury). Except for mercury, the sampling and analysis methods for most of these species had been implemented by 1993. Mercury monitoring was added at each IADN site by 1995.
- ***Third priority group:*** Compounds such as toxaphene, dioxins/furans, and agrochemicals which have an important atmospheric component but require additional methods development to accurately measure their concentrations in atmospheric deposition samples.

Target compounds were chosen for IADN based on their potential to bioaccumulate, their tendency to be transported atmospherically, and the availability and efficiency of detection methods (see sidebar). A major benefit of IADN is the ability to monitor long-term atmospheric concentration changes of such compounds as PCBs and other chemicals of concern. In the past, such regional-scale atmospheric data have been sparse.

It was originally projected that, to be representative of regional deposition patterns, the IADN required one "master" station and several "satellite" stations on each lake. The master stations are: Eagle Harbor, MI; Sleeping Bear Dunes, MI; Burnt Island, Ontario; Sturgeon Point, NY; and Pt. Petre, Ontario (see Figure IV-5). Several satellite sites were later added, including an urban site in Chicago (see box on next page). The IADN implementation design allows for periodic evaluation of the existing sites to determine whether other sites are needed.

At each IADN site, concentrations of target chemicals are measured in rain and snow (wet deposition), airborne particles (dry deposition), and airborne organic vapors. In addition, precipitation rate, temperature, relative humidity, wind speed and direction, and solar radiation are measured at each site. IADN results for selected pollutants are presented in Tables IV-1 and IV-2.

At this time, after five years of operation, many of the sampling and analysis issues of IADN have been resolved. For example, comparability of sampling and analytical procedures between jurisdictions was achieved through extensive laboratory intercomparison studies. However, the uncertainty in the analytical measurement of some compounds is still above the uncertainty threshold acceptable to most policy makers. Toxic chemicals at extremely low concentrations in air, such as PCBs and some agricultural chemicals, have the highest uncertainties in sampling (over 40 percent). Relative standard deviations of air concentrations of organochlorinated compounds may vary from 60 to 90 percent, due to seasonal and annual fluctuations in the air, rather than precision of the measurement (Hoff et al. 1996).

Uncertainty in the deposition estimates may result from various factors: (1) general approximations for estimating deposition; (2) climatic and meteorological variations; (3) differences in the instrumentation and the scope and objectives of the various jurisdictions and agencies involved; and (4) estimation of factors used to calculate loadings (e.g., magnitude of Henry's Law constants, rates of contaminant transfer between the air and water). Despite these

Urban Influence on Atmospheric Deposition of Contaminants

The primary focus of IADN is to determine regionally representative atmospheric deposition loadings of toxic chemicals to the Great Lakes. Thus, monitoring stations were positioned to minimize the influence of local sources and to monitor the atmospheric environment over the lakes as much as possible. This approach does not directly enable the determination of the role of urban air pollution. Recent research suggests that deposition of contaminated large particles carried by winds passing over urban areas can result in substantial inputs of toxic chemicals to the Great Lakes (Falconer et al. 1995; Holsen et al. 1991).

The influence of pollution from the Chicago-northwest Indiana area on water quality in southern Lake Michigan was studied by Sweet and Basu (1994). The Sleeping Bear Dunes site (in the State of Michigan) is located one kilometer from the northeastern shore of Lake Michigan and 50 kilometers from the nearest urban area or major source and, thus, is considered a "remote" site. The first urban site is located 1.5 kilometers from the shore on the campus of the Illinois Institute of Technology, which is near major expressways and surrounded by commercial and residential areas. The second urban site is located at the Indiana Dunes National Lakeshore in the vicinity of large steel mills. Particulate concentrations were measured for target compounds (PCBs, pesticides, and trace metals). Gas concentrations of PCBs and pesticides were determined, and rain was analyzed only for PCBs.

Results from Sweet and Basu (1994) indicate that, for PCBs, DDT (and its metabolites), dieldrin, chlordane, and several trace metals (manganese, zinc, chromium, and lead), the measured particulate and gas concentration values were 10 to 40 times higher in urban areas than at the remote site. For other pesticides (α-HCH, lindane, HCB) and trace metals (arsenic and selenium), concentrations were nearly the same at all three sites, indicating these pollutants were well mixed in the air throughout the region (and that there were probably few local sources).

Though 90 to 99 percent of the PCBs were found in the gas phase, the most toxic PCB congeners were enriched in the particulate phase (Falconer et al. 1995; Holsen et al. 1991; Sweet and Basu 1994). Thus, dry deposition may be an important transport mechanism for certain, especially toxic, PCBs to the lakes. Urban particulate matter also carried high concentrations of trace metals and pesticides, causing dry deposition of these materials to southern Lake Michigan. Dry deposition of large particles may be especially significant for Lake Michigan because 200 kilometers of the southwest shoreline are heavily developed. Prevailing southwest winds carry emissions over the lake where they travel for 100 to 150 kilometers before reaching land again, allowing a significant portion of the deposition to enter the lake. Finally, the concentration of PCBs in precipitation is roughly the same in urban and rural sites. The relatively low levels found in urban precipitation may be due to the fact that many contaminants are collected in clouds or by rain upwind of polluted areas.

Clearly the influence of urban areas on atmospheric deposition of certain pollutants to the Great Lakes is substantial, especially in heavily developed areas, such as the southwestern shores of Lake Michigan.

limitations, the reported estimates are the best that are currently available. Also, data on the concentration of contaminants in the water column for all the Great Lakes have improved recently with more samples being collected and analyzed.

GREAT LAKES EMISSIONS INVENTORIES

A significant step toward assessing the need to reduce atmospheric loads of hazardous air pollutants to the Great Lakes is to identify, categorize, and estimate the magnitude of the pollutant sources. By creating an emissions inventory data base, it is possible to identify the sources and source categories that contribute most to the total emissions in a given geographic area, as well as to model emissions transport and deposition. An air emissions inventory is typically based on mathematical estimates of pollutant releases through the use of emission factors (e.g., a number that represents emissions per unit burned, produced, or processed). These emission factors are derived from actual measurements of the emissions from representative sources and are derived specifically for one type of process or process equipment. Emission factors can be used, for example, to estimate both the amount and type of pollutants being emitted from an air pollution source based upon the quantities of material processed.

The 1986 Great Lakes Governors' Toxic Substances Control Agreement specified provisions to address atmospheric deposition, including a commitment "to cooperate in quantifying the loadings of toxic substances originating from all sources, with the purpose of developing the most environmentally and economically sound control programs." In response to the governors' direction, the air regulatory agencies in the eight Great Lakes states and the province of Ontario began to work cooperatively in 1987 to investigate "the development of a computerized air toxics data base for the purpose of obtaining a better understanding of the nature and sources of toxic air emissions and their migration, dispersion, and resulting impact upon the Great Lakes basin." Under the auspices and management of the Great Lakes Commission (representing the eight Great Lakes states) and with major funding from EPA's Great Waters program, the Great Lakes states began developing a *regional* air toxics emissions inventory. This first regional inventory is scheduled for completion in 1997 and is expected to compile 1993 emissions data for 49 toxic air pollutants from point and area sources. Emissions data on toxic air pollutants from mobile sources will be developed in 1997-1998. These 49 pollutants include 10 of the Great Waters pollutants of concern (cadmium, chlordane, hexachlorobenzene, lead/alkylated lead, mercury, PCBs, PAHs, POMs, TCDD, TCDF). The continued partnership of the region's air regulatory agencies, now in its eighth year, and the high level of regional cooperation and coordination exemplifies the commitment to decreasing toxic deposition into the Great Lakes ecosystem. Yet the inventory must be accompanied by an ongoing commitment to further quantify, assess, and report on the effects of voluntary and regulatory reductions of air toxics emissions.

The key to the state's coordinated efforts is *The Air Toxics Emissions Inventory Protocol for the Great Lakes States*, developed in June 1994 (see sidebar). The Protocol will be followed by each participating state ensuring that consistent, agreed-upon best methodologies are used among all states when compiling a quality-assured inventory. This Protocol is an evolving document and will be updated or revised as needed and agreed upon by all the Great Lakes states.

The second fundamental component in developing a toxic air emissions inventory is the Regional Air Pollutant Inventory Development System (RAPIDS), a multi-state pollutant emissions estimation and storage software system. RAPIDS is a state-of-the-art, networked, relational data management and emission estimation

Components of Great Lakes Emissions Inventory

To date, the Great Lakes States have developed and tested two fundamental components of the inventory effort:

1. ***The Air Toxics Emissions Inventory Protocol for the Great Lakes*** - A guide for each state's efforts to identify sources and estimate emissions so that the inventory is complete, accurate, and consistent from state to state.

2. **RAPIDS** - A client/server relational database software and data management and emissions estimation system. It was designed so that the Great Lakes states may adopt RAPIDS (or some variant of it) for their state system, and may also submit their data for incorporation with the regional RAPIDS data base at EPA.

system, bridging each state's individual inventory and computer system to the regional RAPIDS repository of inventory data. RAPIDS' strength is its versatility. States can modify or build upon it to serve their particular needs. It can be used to estimate both toxic and criteria pollutant emissions from a single device within a facility or a complex grouping of devices and controls, or even across geographic areas, ranging in size from one facility to the entire Great Lakes region. It is designed to run on a personal computer and applies a flexible data model that can be easily expanded in the future to support multi-media, permitting, monitoring, reporting, and compliance activities in the states. Emission factors are uploaded from EPA's Factor Information Retrieval System (FIRE), which contains quality-rated emission factors for both criteria and hazardous air pollutants.

Using RAPIDS, the Great Lakes states' air regulatory agencies are building a comprehensive, updatable statewide and regional air toxics inventory for point, area, and (in the future) mobile sources for the 49 air pollutants. Each of the eight Great Lakes states will be responsible for compiling, uploading, and validating their state emissions inventory data. It is anticipated that the inventories will be updated on a one- or two-year basis.

Four states (Illinois, Indiana, Wisconsin, and Michigan) completed a pilot study of major urban areas along the southwest shore of Lake Michigan in December 1995 using RAPIDS and the Protocol. The states created an inventory of small point and area source categories in the 12 shoreline counties encompassing Chicago (Illinois), Gary (Indiana), and Milwaukee (Wisconsin) that contribute the most to the total emissions of the 49 pollutants of concern. These area sources include gasoline stations, foundries, asphalt and cement plants, and hospitals, among others. The project was the first rigorous test of the regional Protocol and the RAPIDS software. Total pollutant emissions from the inventoried sources were collected, but data interpretations and conclusions were not developed from the results. Instead, the process of compiling the regional inventory was used as a means of resolving many technical, methodological, and policy-related issues that impact a multistate, regional toxic air emissions inventory. Furthermore, the pilot study provided useful information on serious shortcomings that still exist in the regional emissions estimates and suggested necessary steps that must be made to ensure data quality for estimating various pollutant groups. The results from this pilot study will also contribute for better methodology for use in the full eight-state regional inventory.

The level of emissions resolution and the source categories contained in RAPIDS were planned to meet the modeling needs of Great Lakes air quality researchers. This inventory will be available for dispersion and deposition models to characterize source, source category, and geographic contributions, and for mass balance models to characterize media contributions.

Toxics Reduction Efforts in the Great Lakes

In recent years, several programs/activities have developed approaches to reduce loadings and to mitigate existing contamination. These programs are described below. They may provide information to further characterize and reduce atmospheric contamination in the Great Lakes region.

VIRTUAL ELIMINATION

The Great Lakes Water Quality Agreement of 1978 between the United States and Canada called for the "virtual elimination" of persistent toxic substances, especially those which bioaccumulate, from the Great Lakes basin. In keeping with the obligations of the Great Lakes Water Quality Agreement, two major efforts have occurred: (1) a pilot project sponsored by EPA to develop the framework to achieve virtual elimination of two pollutants, mercury and PCBs; and (2) development of the Great Lakes Binational Toxics Strategy (released in April 1997) between the United States and Canada to set goals to reduce the use and release of selected pollutants. Many of the recommendations from the pilot project were incorporated into the Binational Toxics Strategy.

Virtual Elimination Pilot Project. Since 1990, both the United States and Canada have initiated separate efforts for virtual elimination. EPA began the Virtual Elimination Pilot Project in 1993, which was designed to answer the following question: "What options exist for improving the current regulatory and non-regulatory framework to encourage continuing reductions towards zero in the use, generation, and release of selected toxic substances?" The aim of the project was to identify barriers to achieving virtual elimination and to develop strategies to overcome these barriers.

The first iteration or "pilot" portion of the U.S. project focused on the reduction opportunities for two substances, mercury and PCBs. EPA held a meeting with stakeholders in the Great Lakes region in 1993 to share information on mercury and PCBs, and to offer participants the opportunity to make recommendations on ways to reduce the use and release of each pollutant. Based on the results of this meeting, a draft report was developed by EPA to identify options to reduce mercury (GLNPO 1995). A draft options paper for the virtual elimination of PCBs is currently being prepared by EPA. It is expected that this project will continue with the analyses of classes of substances rather than the use of a chemical-by-chemical approach.

Mercury presents an unusual challenge to society because of its semi-volatility, persistence, complex environmental chemistry, and tendency to bioaccumulate in fish. The draft mercury options paper (GLNPO 1995) proposed regulatory and voluntary measures to prevent or reduce atmospheric mercury contamination, and introduced the concept of the mercury "life cycle." A comprehensive approach to virtually eliminating mercury releases was proposed:

- Increase public awareness of mercury problems and mercury-containing items;
- Influence supply of mercury to minimize primary production and manage federal holdings;
- Minimize use of mercury through pollution prevention and alternative technologies;
- Reduce uncontrolled releases by encouraging recycling and regulating releases; and
- Manage disposal of mercury-containing items and mercuric wastes.

Regulatory and Voluntary Options that may Prevent or Reduce Atmospheric Mercury Contamination

- ***Pollution Prevention.*** Mercury use in the United States has substantially declined during the past two decades. This trend parallels that of western Europe. Ebbing use implies that less mercury will be included in waste streams reaching incinerators and landfills, or released during production processes. The decline in use is attributable to both government bans and technological advances that create environmentally safer products. Mercury has been banned in pesticides (1972-1976 and 1993) and in paints. The number of U.S. chloralkali facilities using a mercury cell process has declined from 25 during the early 1970s to 14 in recent years. Minnesota, Wisconsin, and New York restricted mercury content in batteries, and consequently, national use of mercury in batteries fell from 448 tons during 1988 to 10 tons during 1993. Mercury content in fluorescent bulbs has also declined.

- ***Recycling.*** A number of users of mercury are taking steps to boost recycling. Various municipal wastewater treatment plants are working with dental associations to encourage recovery of mercury in dentist offices so that mercury amalgam does not enter their treatment systems. Some manufacturers who rely on mercury in instruments are starting or considering take-back programs; EPA and states are working with several such firms to ensure that liability concerns do not preclude voluntary efforts. A number of hospitals are diverting mercury-containing wastes from incineration. Several states require that fluorescent bulbs be recycled (e.g., Florida, Minnesota), because such bulbs are commonly broken in or en route to landfills, allowing mercury vapors to escape to the atmosphere. While the quantity of mercury contained in a single bulb is minuscule, the number of discarded bulbs is great.

- ***Management.*** Pursuant to implementation of the CAA, EPA has proposed performance standards for municipal and medical waste incinerators. A final rule for municipal waste combustors (new sources with individual capacity of >250 tons/day) was promulgated in December 1995 (60 *Federal Register* 65387) and a proposed rule for medical waste incinerators in June 1996 (61 *Federal Register* 31736). There are about 2,400 medical and 180 municipal incinerators across the nation. These sectors are estimated to generate about one-half of current national emissions of mercury, primarily through combustion of discarded products that contain mercury. When fully implemented by 2002 through state plans, these proposed standards hold promise of reducing mercury emissions from these two classes of incinerators by 95 and 80 percent, respectively. Performance standards will also be evaluated for other sectors that also emit mercury, including the Portland Cement industry; commercial and industrial boilers; primary lead smelters; the chloralkali sector; primary copper smelters; sewage sludge incinerators; and lime manufacturing.

- ***Global Production and Release of Mercury.*** During recent years, the U.S. government has held a significant stockpile of mercury on the world commodity market. To dispose of its holding, the Department of Defense has periodically auctioned mercury. From 1988 through 1993, sales totaled four million pounds. Sales were suspended in 1994, pending consideration of their environmental impacts. One positive environmental impact of sales is that they may forestall virgin production of mercury; the last mercury mines in the United States closed several years ago. Both western Europe and the United States have become substantial net exporters as their mercury consumption has fallen, whereas world use may be growing (Lawrence 1994). Because of diminished use of mercury, the United States is nearly meeting its entire need for mercury through recycling. Much of the mercury sold by the federal government has been exported. Foreign use and release, due to less stringent controls, can contribute to global atmospheric contamination which can travel for long distances, and directly contaminate U.S. surface waters. Suspension of government sales has positioned the United States to seek needed international cooperation in minimizing mercury releases on a global scale.

The Great Lakes Binational Toxics Strategy. The *Canada-United States Strategy for the Virtual Elimination of Persistent Toxic Substances in the Great Lakes*, also known as the Binational Great Lakes Toxics Strategy, was signed between the two countries on April 7, 1997 (U.S. EPA and Environment Canada 1997). This Binational Strategy was developed jointly by EPA and Environment Canada, in keeping with the objectives of the 1987 GLWQA. Both Canada and

United States have domestic virtual elimination strategies but a coordinated strategy is necessary for the greatest reduction in toxic substances throughout the Basin. Both nations encourage and support voluntary programs by all stakeholders to reduce the generation, use, and release of toxic substances to the Great Lakes.

A Four-Step Process Toward Virtual Elimination

1. Gather information on generation, uses, and sources of the pollutant within and outside the Great Lakes Basin;
2. Analyze current regulatory and non-regulatory programs and initiatives that manage or control the pollutants and identify the gaps in these regulations that offer opportunities for reductions;
3. Develop cost-effective options and provide recommendations for increasing the pace and level of reductions; and
4. Recommend and implement actions to achieve goal.

The Binational Strategy provides the framework to achieve quantifiable goals in a specified time frame (1997 to 2006) for targeted persistent toxic substances, especially those which bioaccumulate. Flexibility is provided in the Strategy to allow for the revision of targets, time frames, and the list of pollutants. All actions and activities, both regulatory and nonregulatory, will be considered to help speed reductions. The Strategy also recommends that goals be accomplished through a four-step process (see sidebar). The pollutants identified in the Strategy fall into two lists. The Level I substances represent an immediate priority and are targeted for reduction and eventual virtual elimination through pollution prevention and other incentive-based actions. These pollutants are aldrin/dieldrin, benzo(a)pyrene, chlordane, DDT/DDE/DDE, hexachlorobenzene, alkyl lead, mercury and mercury compounds, mirex, octachlorostyrene, PCBs, dioxins/furans, and toxaphene. For pollutants that are considered Level II substances, the governments encourage stakeholders to undertake pollution prevention activities to reduce levels in the environment of those substances nominated jointly by both countries, and to conform with the laws and policies of each country, including pollution prevention, with respect to those substances nominated by only one country, until and unless these pollutants are placed on the Level I list. The Level II pollutants are cadmium and cadmium compounds, 1,4-dichlorobenzene, 3,3'-dichlorobenzidine, dinitropyrene, endrin, heptachlor (and heptachlor epoxide), hexachlorobutadiene (and hexachloro-1,3-butadiene), hexachlorocyclohexane, 4,4'-methylenebis(2-chloroaniline), PAHs, pentachlorobenzene, pentachlorophenol, tetrachlorobenzene (1,2,3,4- and 1,2,4,5-), and tributyl tin.

Both the United States and Canada have set "challenge" goals to achieve reductions through implementation of voluntary efforts and regulatory actions. One of these challenges is the commitment of these countries to work together to assess atmospheric inputs of persistent toxic substances to the Great Lakes, with the goal of evaluating and reporting jointly on the contribution and significance of long-range transport of these substances from worldwide sources. Efforts will be made to work within the existing international framework to reduce releases of such pollutants from remaining long-range sources. Activities by EPA and Environment Canada to meet this particular challenge include:

- Coordinate efforts to identify sources in order to better define and coordinate emission control programs;

- Maintain atmospheric deposition monitoring stations to detect deposition and transport of toxic substances;
- Continue research on atmospheric science of toxic pollutants to refine and to improve existing source, receptor, and deposition models, as well as improve integration of existing air toxic monitoring networks and data management systems to track deposition of contaminants within the Great Lakes; and
- Conduct an assessment of long-range transport of persistent toxic pollutants from worldwide sources

In addition to these coordinated binational efforts, Environment Canada will also demonstrate alternative processes to lessen emissions from five predominant sources by 2001 and complete inventories of 10 selected air pollution sources to support assessment of environmental impacts of air toxics by 1999.

Besides the above challenge, the Strategy includes several specific reduction goals or challenges for the Level I pollutants (Table IV-3). For the United States, the baseline from which these reductions will be measured will be the most recent and appropriate inventory (e.g., mercury will be based on estimated emissions during the early 1990s). Canada plans to use their 1988 emissions inventory.

Two additional challenges from the Strategy are: (1) complete or be well advanced in remediation of priority sites with contaminated bottom sediments in the Great Lakes Basin by 2006; and (2) promote pollution prevention and sound management of Level II substances, to reduce levels in the environment. The Binational Strategy is intended to fill in the gaps that exist where ongoing programs or emerging initiatives do not address toxic releases, to provide a context of basinwide goals for localized actions, and to provide "out of basin" support to programs such as LaMPs.

LAKEWIDE MANAGEMENT PLANS (LAMPS)

In Article VI, Annex 2 of the GLWQA, the U.S. and Canadian governments agreed to develop and implement LaMPs for each of the five Great Lakes. The purpose of the LaMPs is to document an approach to reducing input of critical pollutants to the Great Lakes and restoring and maintaining Great Lakes integrity. LaMPs are management tools designed to (1) integrate federal, state, provincial, and local programs to reduce loadings of toxic substances from both point and nonpoint sources; (2) assess whether these programs will ensure attainment of water quality standards and designated beneficial uses; and (3) recommend any media-specific program actions or enhancements to reduce toxic loadings in waters currently not attaining water quality standards and/or designated beneficial uses. Unlike the other four Lakes, Lake Michigan lies entirely within the boundaries of the United States and therefore, the Lake Michigan LaMP has been developed solely by U.S. federal and state agencies with input from a public forum. The development of this program, as well as the deadlines established for the completion of the program, is mandated under section 118 of the CWA. In addition, as noted in Chapter I, section 112(m) of the CAA requires that EPA, in cooperation with NOAA, monitor the Great Lakes, investigate atmospheric deposition rates and pollutant sources, improve monitoring methods, and determine the relative contribution of atmospheric pollutants to the total pollution loadings to the Great Lakes and other Great Waters.

TABLE IV-3
Specific Pollutant Reduction Goals Under the Great Lakes Binational Toxics Strategy [a]

Level I Substances	United States Challenge	Canadian Challenge
Aldrin/Dieldrin, Chlordane, DDT, Toxaphene, Mirex, Octachlorostyrene	Confirm by 1998 that there is no longer use or release from sources that enter Great Lakes Basin. If ongoing long-range sources from outside of U.S. are confirmed, use existing international frameworks to reduce or phase out releases.	Report by 1997 that there is no longer use, generation, or release from Ontario sources that enter Great Lakes Basin. If ongoing long-range sources outside of Canada are confirmed, use existing international frameworks to reduce or phase out releases.
Alkyl lead	Confirm by 1998, there is no longer use in automotive gasoline; support and encourage stakeholder efforts to reduce releases from other sources.	Seek by 2000, 90 percent reduction in use, generation, or release.
PCBs	Seek by 2006, a 90 percent reduction nationally of high level PCBs (>500 ppm) in electrical equipment; ensure all PCBs retired from use are properly managed and disposed of to prevent accidental releases within or to the Great Lakes Basin.	Seek by 2000, a 90 percent reduction nationally of high-level PCBs (>1 percent PCBs) that were once, or are currently, in service and accelerate destruction of stored high-level PCB wastes that may enter the Great Lakes Basin.
Mercury[b]	Seek by 2006, a 50 percent reduction nationally in deliberate use of mercury and a 50 percent reduction in release from sources resulting from human activity.[c]	Seek by 2000, a 90 percent reduction nationally in releases of mercury, or where warranted the use of mercury, from polluting sources resulting from human activity in the Great Lakes Basin.
Dioxins/Furans	Seek by 2006, a 75 percent reduction in total releases of dioxins/furans (2,3,7,8-TCDD toxicity equivalents) from sources resulting from human activity.[c]	Seek by 2000, a 90 percent reduction in releases of dioxins/furans (focus on 2,3,7,8-substitute congeners) from sources resulting from human activity in Great Lakes Basin.
Benzo(a)pyrene, HCB	Seek by 2006, reductions in releases that are within or may have potential to enter the Great Lakes Basin from sources resulting from human activity.	Seek by 2000, a 90 percent reduction in releases from sources resulting from human activity in the Great Lakes Basin.

[a] Detailed descriptions of these challenges are presented in The Great Lakes Binational Toxics Strategy (U.S. EPA and Environment Canada 1997).
[b] Mercury challenges are considered interim reduction targets for mercury and, in consultation with stakeholders, will be revised if warranted, following completion of EPA's Mercury Study Report to Congress (U.S. challenge) and 1997 *Canada-Ontario Agreement Respecting the Great Lakes Basin Ecosystem* (Canadian challenge).
[c] The release challenge applies to the aggregate of releases to the air nationwide and of releases to the water within the Great Lakes Basin.

A LaMP is a dynamic, action-oriented process encompassing a number of components. These include an evaluation of beneficial use impairments and pollutants contributing to those impairments; a summary of sources and loads of these critical pollutants; identification of ongoing prevention, control, and remediation actions, as well as additional efforts needed to reduce pollutant loads and to restore beneficial uses; and monitoring activities to evaluate the effectiveness of program actions. This approach for developing and implementing LaMPs is an evolutionary and iterative process for identifying and reducing critical pollutants. Public participation and cooperation with states and local governments is a key component to the LaMP development process.

LaMPs are in various stages of development for each of the Great Lakes (see sidebar). Not all of the Lakes have LaMPs published in the Federal Register; however, commitments have been made by key stakeholders in the respective basins to pursue toxics reduction. Actions are being taken to achieve this goal. Each LaMP addresses a different list of critical pollutants, but some common ones are mercury, PCBs, hexachlorobenzene, dioxins, furans, chlordane, DDT and metabolites, and dieldrin (all of which are Great Waters pollutants of concern).

Current Status of LaMPs in the Great Lakes

Superior	Binational Program to Restore and Protect the Lake Superior Basin announced (1991) Stage 1 LaMP submitted to IJC (1995) Stage 2 LaMP released for public review (1996)
Michigan	LaMP published in Federal Register (1994)
Huron	LaMP not established
Erie	LaMP Management Committee formed (1994)
Ontario	Lake Ontario Toxics Management Plan (1989) LaMP Workplan signed (1993)

Several activities have been initiated through LaMPs:

- In the Lake Michigan basin, agricultural "clean sweeps" to properly collect and dispose of unused pesticides have been conducted in Indiana, Michigan, and Wisconsin. Also, a variety of pollution prevention and technical assistance projects have taken place in Milwaukee, Chicago, and western Michigan.

- In most lake basins, tributary and atmospheric deposition monitoring is occurring. The Lake Michigan LaMP is utilizing the information generated from the Lake Michigan Mass Balance Study (described earlier in the section) to identify and reduce loadings as data become available.

- The Lake Superior LaMP was initiated as a component of the binational efforts to restore and protect Lake Superior. One of the goals of the LaMP is to achieve zero discharge and emission of persistent toxic pollutants through its Zero Discharge Demonstration Project. Another effort of the Lake Superior LaMP is an extensive pollution prevention outreach and education program developed for mercury. Among the activities are battery collection, energy efficiency, and product takeback programs. For example, Honeywell, Inc., the largest manufacturer of mercury thermostats used in regulating heating in the home, has instituted a thermostat takeback program in which the company recycles the mercury.

The LaMPs often provide the needed coordination and oversight for many such projects being implemented all over the Great Lakes Basin.

THE GREAT LAKES WATER QUALITY (GLWQ) GUIDANCE

Another notable toxics reduction effort was the GLWQ Guidance (U.S. EPA 1995a). It is the culmination of a six-year cooperative effort that included participation by the eight Great

Lakes states, the environmental community, academia, industry, municipalities, and EPA's regional and national offices, and stems from the Great Lakes Water Quality Initiative, which began when the states of the Great Lakes Region recognized the important feature of the Great Lakes ecosystem to accumulate persistent pollutants. The guidance is not only designed to address existing problems, but also to prevent emerging and potential problems posed by additional chemicals in the future, which may damage the overall health of the Great Lakes system. The guidance includes criteria for the protection of human life, wildlife, and aquatic life, taking into account the ability of many pollutants to biomagnify. Antidegradation requirements assure that current water quality will not be diminished. The guidance also outlines procedures to ensure consistent implementation and appropriate flexibility for long-term protection of the Great Lakes.

The GLWQ Guidance promotes the use of pollutant minimization plans to stop pollution before it enters the environment. Reducing pollution at its source is the most effective way of protecting public health and the environment, and is often more economical than cleaning up after a pollutant is released.

The water quality criteria in the GLWQ Guidance apply to all of the waters in the Great Lakes system, regardless of the source of pollution. Pollutants enter the Great Lakes from the air, stirred-up bottom sediments, urban and agricultural runoff, hazardous waste and Superfund sites, spills, and industrial and municipal wastewater discharges. Although the implementation procedures of the guidance apply mostly to industrial and municipal water discharges, a state may find it is more effective (or cost-effective) to improve water quality by reducing air emissions or cleaning up contaminated sediments, and may choose not to impose additional requirements on wastewater dischargers.

GREAT LAKES REMEDIAL ACTION PLANS

The 1978 GLWQA, along with the 1987 amendments, established guidelines for restoring the quality of the Great Lakes. As a response to this measure, geographical "problem areas" or Areas of Concern (AOCs) were identified in the Great Lakes where GLWQA objectives had been exceeded and such exceedance had caused, or was likely to cause, impairment of beneficial use or the area's ability to support aquatic life. The defined AOCs include rivers, connecting channels, harbors, and embayments of the Great Lakes, with the U.S. states and Canadian provinces responsible for remediating these areas. The Water Quality Board (WQB) of the IJC determines the AOCs, but the specific geographical boundaries of the AOCs are set by the states and/or provincial governments. Currently, there are 43 AOCs; 12 are under Canadian jurisdiction, 26 under U.S. jurisdiction, and five under jurisdiction of both countries. The sources of contamination have usually been water discharges from point and non-point sources.

In order to provide more uniform guidance on how to remediate the AOCs, Remedial Action Plans (RAPs) were introduced in 1985. In 1994, eight U.S. states were involved in the RAP process: Indiana, Illinois, Minnesota and Pennsylvania (1 AOC each), Ohio (4 AOCs), Wisconsin (5 AOCs), New York (6 AOCs), and Michigan (14 AOCs). Currently, for most AOCs, the problem definition stage of the RAP process has been addressed and the planning and implementation stages are to be initiated. One of the major problems facing the AOCs today is toxic contamination of the sediments. All of the U.S. AOCs have impaired beneficial uses attributable to contaminated sediments (U.S. EPA and Environment Canada 1994). As a result, sediment remediation is a key component of RAP remediation.

The progress of AOC remediation is presented in an EPA report, *Progress in the Great Lakes Remedial Action Plans: Implementing the Ecosystem Approach in the Great Lakes Areas of Concern* (U.S. EPA and Environment Canada 1994). This report provides an update for each AOC in the Great Lakes and summarizes the major barriers to and benefits of the RAP process. It concludes that a comprehensive decision-making process that leads to commitment for action is an essential aspect of implementing an ecosystem approach, as required in a RAP. Great Lakes federal, state, and provincial governments have provided leadership and resources for development and implementation of RAPs. Through government and community-based partnerships, RAPs are being developed to be a coordinated, multi-stakeholder response to restoring impaired beneficial uses in AOCs.

SOME ADDITIONAL ACTIONS RELATED TO TOXIC CONTAMINATION AND REDUCTION

In addition to activities described above, many pollutant-specific efforts provide significant information on atmospheric contamination in the Great Lakes. For example, EPA developed standards for municipal waste combustors (excludes new sources with individual capacity of $\leq$250 tons/year) (60 *Federal Register* 65387) and proposed standards for medical waste incinerators (61 *Federal Register* 31736) which will, when implemented by 2002, provide about a 70 ton reduction in mercury emissions, or 35 percent of current total U.S. emissions, based on 1990 emissions estimates. Implementation of other Maximum Available Control Technology (MACT) standards, including those proposed in 1996 for hazardous waste combustors, offer the probability of further mercury emission reductions in the future.

For toxaphene, many issues remain about its sources and continuing presence in the Great Lakes. EPA held a research workshop on March 27-29, 1996, with scientists to assess these concerns (U.S. EPA 1996c). Based on the meeting, the following actions were recommended to maintain progress toward resolving several questions concerning concentrations and trends (spatial and temporal) in various Great Lakes media and the sources of toxaphene in the Great Lakes:

- Measure concentration gradient of toxaphene across air-water interface and link with atmospheric source profile to establish seasonal and annual fluxes to and from Lakes Superior and Michigan, as well as correlative measurements in other Great lakes to permit spatial comparisons;

- Collect additional sediment cores in Lakes Superior and Michigan to determine concentrations, accumulation rates, and inventories to help establish past dynamics of toxaphene and assist in efforts to forecast future conditions in the Great Lakes;

- Quantify aquatic food web dynamics to establish how food web influences spatial and temporal variations in toxaphene concentrations in biota.

- Measure physical-chemical properties of toxaphene homologs and congeners.

Besides providing information on sediment deposition and cycling of toxaphene in the Great Lakes, it is anticipated that results from these recommendations would provide a more firm technical basis upon which to explore the need for and extent of appropriate management actions.

Another major binational effort to broadly address Great Lakes issues is the State of the Lakes Ecosystem Conference (SOLEC) organized by the governments of the United States and Canada. In the first SOLEC in 1994, a report (and five background papers) was released on the current condition of the Great Lakes (Environment Canada and U.S. EPA 1995). The report addressed the entire Great Lakes system in terms of ecological and human health, and the stressors which affect it. The six areas of discussion were human health; aquatic community health; aquatic habitat; toxic contaminants; nutrients; and the economy. Although the report did not describe or evaluate pollution control or natural resource management programs, it did focus on environmental conditions within the Great Lakes system. The SOLEC report indicated that there has been considerable improvement in all the Great Lakes compared to 30 years ago, although serious losses in habitat for native plants and animals continues. Nutrient and toxic contaminant concentrations appear to be decreasing, although bioaccumulative pollutants still cause problems. The report presented a "mixed picture" of the current conditions of the Great Lakes and challenges managers and decision makers throughout the basin to obtain adequate information, deal with subtle effects of long-term exposure to low levels of toxic contaminants, protect biodiversity, restore habitat for native plants and animals, connect decisions with ecosystem results, and attain sustainability.

State of the Lakes Ecosystem Conference (SOLEC)

As part of the continuing response by the governments of the United States and Canada to the binational Great Lakes Water Agreement, SOLEC was initiated and held in October 1994. The second meeting of SOLEC was held in Windsor, Ontario, on November 6-8, 1996. SOLEC is viewed as part of a process of sharing information needed to make well informed decisions that affect the ecosystem of the Great Lakes. It is attended by managers and other decision makers from the private sector, and government and non-government environmental organizations.

Building on the findings of the first SOLEC, the 1996 SOLEC (Environment Canada and U.S. EPA 1996) focused on nearshore areas of the Great Lakes basin (i.e., warm and shallow waters, coastal wetlands). These areas represent the most diverse and productive parts of the Great Lakes ecosystem, and provide support for most intense human activity and subsequently, are subjected to greatest stress. Key themes from the conference were immediate actions; local/community level involvement; development of common ecosystem health indicators to measure progress; essentiality of cooperation and partnership due to complexity of issues and development of new ideas; approaches that recognize long-term perspective; and focus on prevention and preservation. The Great Waters program will benefit from many aspects of SOLEC, such as the information gathered on the impact of air pollutants on human health and ecological effects and the promotion of pollution prevention measures.

Addressing Data Gaps/Future Needs

Considerable progress has been made in the recent past in characterizing and reducing toxic pollution in the Great Lakes. The programs presented above and summarized in Table IV-4 provide an overview of some notable and recent activities by the United States, as well as Canada, to respond to concerns related to atmospheric pollution in the Great Lakes. Some of these measures include identifying emission sources; characterizing contamination from

TABLE IV-4
Summary of Some Major Programs to Address Atmospheric Contamination in the Great Lakes

Data Collection/Research Projects	
Lake Michigan Monitoring Program/Lake Michigan Mass Balance Study	Scientific base for future load reduction effort at all government levels. Mass balance study addresses CAA section 112(m) through coordinated effort to quantify and understand loadings, transport, and fate of selected HAPs. Also to provide a validated method to estimate loading for other waterbodies.
Integrated Atmospheric Deposition Network (IADN)	Binational monitoring network and research program to determine magnitude and trends of atmospheric deposition for the region.
Great Lakes Emissions Inventories	Inventory of sources and source category emissions in Great Lakes region, with a multi-state data base (RAPIDS).
Toxics Reduction Efforts	
Virtual Elimination Pilot Program/ Great Lakes Binational Toxics Strategy	Regulatory and non-regulatory efforts to encourage reduction in use and release of bioaccumulative pollutants in Great Lakes.
Lakewide Management Plan (LaMP)	Management tool to document approach to decrease pollutant input to each of the Great Lakes.
Great Lakes Water Quality (GLWQ) Guidance	Promotion of pollution minimization plans to stop pollution before it reaches the environment, and consistent standards to protect human health, wildlife, and aquatic life.
Remedial Action Plans for Great Lakes Areas of Concern (AOCs)	Action-planning process for implementing remedial and preventive actions to restore impaired beneficial uses of specific areas.

pollutants; developing and implementing voluntary and regulatory measures; and developing guidance for evaluating levels of risk of pollutant contamination. These programs have usually involved coordination among various federal, state, and/or local agencies. Though each program is designed to address specific goals, many of these programs coordinate their efforts to ensure that results are not duplicated. For example, the Binational Virtual Elimination Strategy was developed to achieve virtual elimination of persistent toxic pollutants in the Great Lakes, but it also supports and builds upon ongoing processes in the LaMPs, such as the Zero Discharge Demonstration Project through the Lake Superior LaMP. Furthermore, the Great Lakes Emissions Inventories will eventually provide information for determining whether the reduction goals set in the Great Lakes Binational Toxics Strategy have been met. The IADN will monitor whether pollutant levels are actually decreasing.

The Great Lakes programs described in this chapter also complement the Great Waters program in assessing and identifying the extent of atmospheric contamination of hazardous air pollutants to the Great Lakes. Further coordinated progress is needed to improve knowledge and understanding of pollutant contamination, as well as to increase public awareness. In addition to the continuation of the current programs/activities, some high priority efforts for the Great Lakes basin include:

- Improvement in research and monitoring techniques to reduce uncertainties in loading calculations (such as those for the IADN) and therefore, result in better estimates of atmospheric pollutant levels and deposition;

- Improvement in dispersion and deposition models currently being developed to link emission inventory information to atmospheric loadings of Great Lakes pollutants at the water's surface;

- After Lake Michigan Mass Balance data have been analyzed, application of results and modeling tools from the study to the development of a general mass balance model for other hazardous air pollutants;

- Increase in efforts to identify local and long-range sources of Great Lakes pollutants through various source apportionment modeling and emissions inventories, such as in the RAPIDS data base; and

- Continuation of efforts to develop and implement strategies and recommendations to reduce use, generation, and release of pollutants affecting the Great Lakes, particularly through binational efforts such as the reduction challenges proposed under the Binational Virtual Elimination Strategy.

As more information is gathered on the characterization and reduction of atmospheric deposition of toxic pollutants to the Great Lakes, the results of these efforts, as well as the tools used, may be applied to other waterbodies, such as Lake Champlain.

IV.B Lake Champlain

Located around the northernmost borders of the states of New York and Vermont and the southern border of the province of Quebec, Lake Champlain is one of the largest freshwater lakes in the United States, with 1,127 km^2 of surface water, over 70 islands, and 945 km of shoreline (see Figure IV-6). It flows north from Whitehall, New York, almost 193 km across the U.S.-Canadian border to its outlet at the Richelieu River in Quebec, where it joins the St. Lawrence River. Lake Champlain is unique because of its narrow width (19 km at its widest point), great depth (over 122 meters in some parts), and large size of the watershed relative to the lake surface.

The Lake Champlain Basin, composed of the entire watershed or drainage area, spans from the Adirondack Mountains in the west to the Green Mountains in the east, and from the Taconic Mountains in the southeast to the St. Lawrence Valley in the north (Figure IV-6). The total area of the Basin is 21,326 km^2, of which 56 percent is in Vermont, 37 percent is in New York, and 7 percent is in Quebec. The Basin is characterized by an 18:1 ratio of watershed to lake surface area, indicating that the lake represents only about 5 percent of the total basin area. Approximately 89 percent of the Basin is categorized as forest and agricultural land.

By the end of 1994, approximately 645,000 people resided in the Lake Champlain Basin, with the population increasing each year by about 1.2 percent (LCBP 1994). Most of the region is classified as rural, with only Burlington, Vermont, recognized as a metropolitan area (1990 population of 52,000). The Lake Champlain Basin has traditionally had a rural resource-based economy, including agriculture, renewable natural resources (e.g., timber, fish, maple syrup), and non-renewable natural resources (e.g., iron ore, marble, gravel). In recent years, the economy has diversified into other activities, but is still dependent on the natural resources (see sidebar).

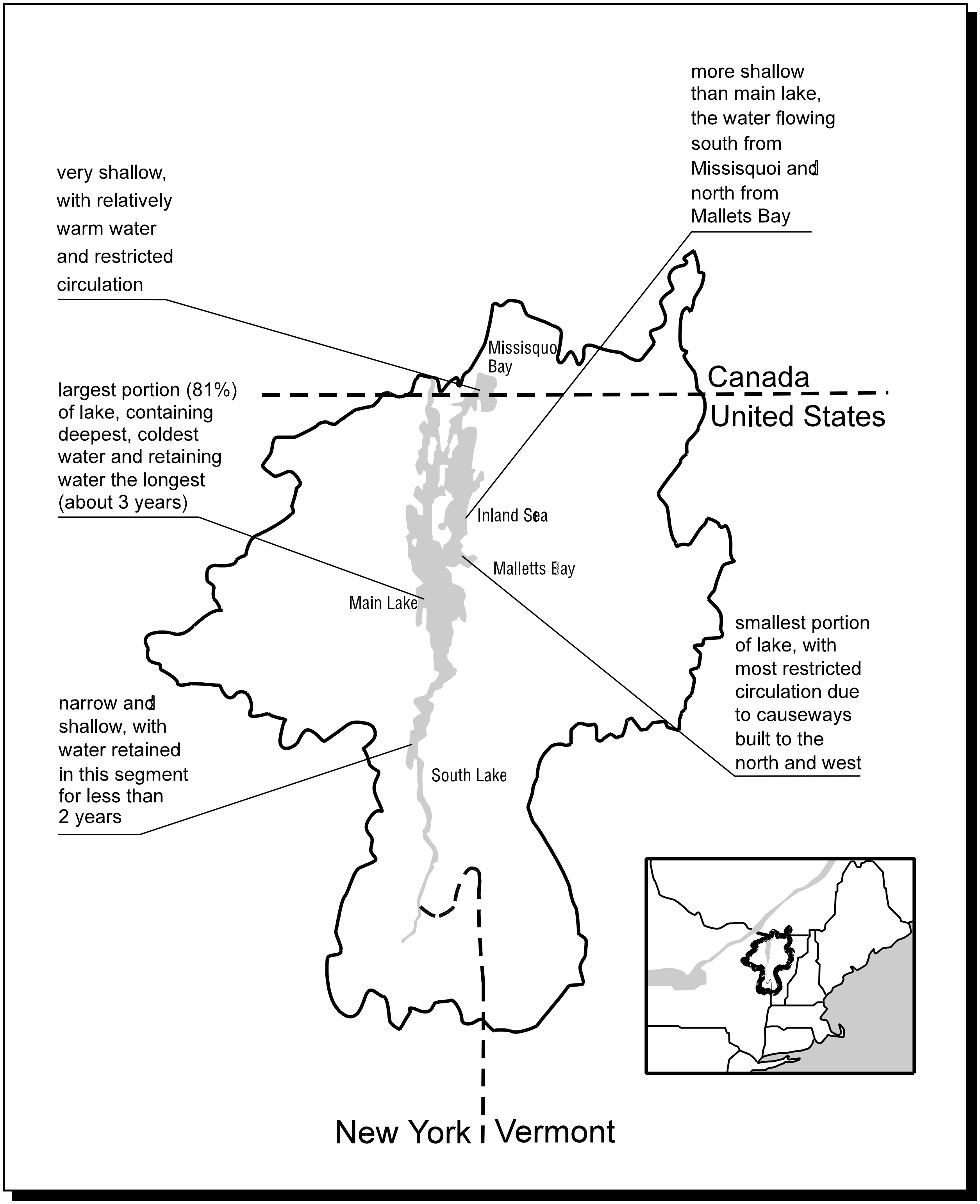
Economic Highlights of Lake Champlain Basin

- Twenty-five percent of the workforce is employed in natural resource-related activities (e.g., agriculture, mining, forestry).
- Resources of the Lake are a major reason why many Basin residents reside in this region.
- Tourism represents a significant economic factor for the region, generating $2.2 billion in 1990, of which 40 percent were Lake related (e.g., marina, white-water rafting). Part of tourism income comes from recreational activities, including $81 million per year from fishing industry (in 1991) and $50 million per year from bird and wildlife viewing (in 1990).

Sources: LCBP 1994, 1996.

Lake Champlain, unlike many other lakes that tend to be more evenly mixed, is made up of five distinct areas or lake segments, each with different physical and chemical characteristics (Figure IV-6). In these lake "divisions," pollutants may accumulate in shallow areas or bays where flushing and water circulation are reduced, and may be deposited at the mouths of rivers where runoff carrying sediment and other pollutants from the watershed is discharged. Human activities also concentrate in many of these areas of the lake, increasing the potential for contamination and exposure.

FIGURE IV-6
Lake Champlain Basin



An understanding of the hydrodynamics of Lake Champlain is essential to predicting how and where pollutants are transported, and where they will end up in the lake. A simplified three-dimensional hydrodynamic transport model, being developed with funding by the Lake Champlain Basin Program (LCBP), will be used as a management tool to determine potential effects of pollutant inputs and other changes to the waterbody (LCBP 1994).

Characterizing Toxic Contaminants in Lake Champlain

Levels of toxic contamination in Lake Champlain are low compared to the Great Lakes; however, concerns for protecting the public health still exist. Fish consumption advisories for two Great Waters pollutants (mercury and PCBs) are currently in effect in both New York and Vermont for fish from Lake Champlain (see Appendix B) (LCBP 1994). Testing of lake bottom sediment near the urbanized sites along the Lake shows pollutants levels that may be of concern and indicates potential risks to aquatic life (see Section II.B). Because of these findings, as well as exceedances of water quality standards set by EPA, NOAA, and the province of Ontario, LCBP gives highest priority to these two pollutants.

The contribution of air deposition as a source of loadings for pollutants, such as metals and organic compounds, is of concern for Lake Champlain and the Basin, and has been the subject of recent studies. Although emissions of toxic pollutants within the Basin are considered low because of the few industries and utilities in the local area, high levels of pollutants may reach the Lake from more distant sources (LCBP 1994, 1996). Data have been limited regarding atmospheric sources, or the movement of pollutants from the atmosphere to the Lake directly or through the watershed.

Atmospheric Sources of Toxic Contamination in Lake Champlain

Local	Mobile emissions Residential energy consumption (e.g., wood burning) Waste incinerators
Regional/ Long-distance	Smelters (Quebec) Utilities (midwestern United States)

A 1990/1991 air monitoring study measured the concentration of toxic metals at four sites in the Lake Champlain Basin (Whiteface Mountain, NY; Willisboro Bay, NY; Burlington, VT; and Underhill, VT) (LCBP 1994). Preliminary data revealed elevated levels of zinc in the air surrounding Burlington, possibly due to refuse incineration, tire wear, and industries. There were also periodic increases in arsenic levels at these and other sites across the Northeast. The source of these arsenic concentrations in the air is believed to be a smelter in Quebec (LCBP 1994). Mercury, lead, and cadmium compounds were also measured in this study; however, there was no indication that the concentrations for these Great Waters pollutants of concern were of concern at the monitoring sites.

The following subsection focuses on currently ongoing mercury research to determine atmospheric deposition to the Lake Champlain Basin. Information on atmospheric deposition of the other high priority pollutant to the basin, PCBs, is lacking at this time.

ATMOSPHERIC DEPOSITION OF MERCURY IN THE LAKE CHAMPLAIN BASIN

Mercury burden in Lake Champlain is evidenced by fish consumption advisories. Direct discharges of mercury are quite limited which has led to concerns that the atmosphere may be the major route of mercury to the lake. Recent studies supported by NOAA and EPA's Great Waters program have investigated the deposition of atmospheric mercury in the Lake Champlain Basin (Burke et al. 1995; Scherbatskoy et al. 1997). The air monitoring data collected from these efforts should provide an initial framework for a more comprehensive analysis of mercury cycling (i.e., deposition, transport, transformation, and accumulation) in the region, and beyond. There is currently no information that compares the estimated amount of mercury entering the lake via water (e.g., runoff) with the amount deposited from the air.

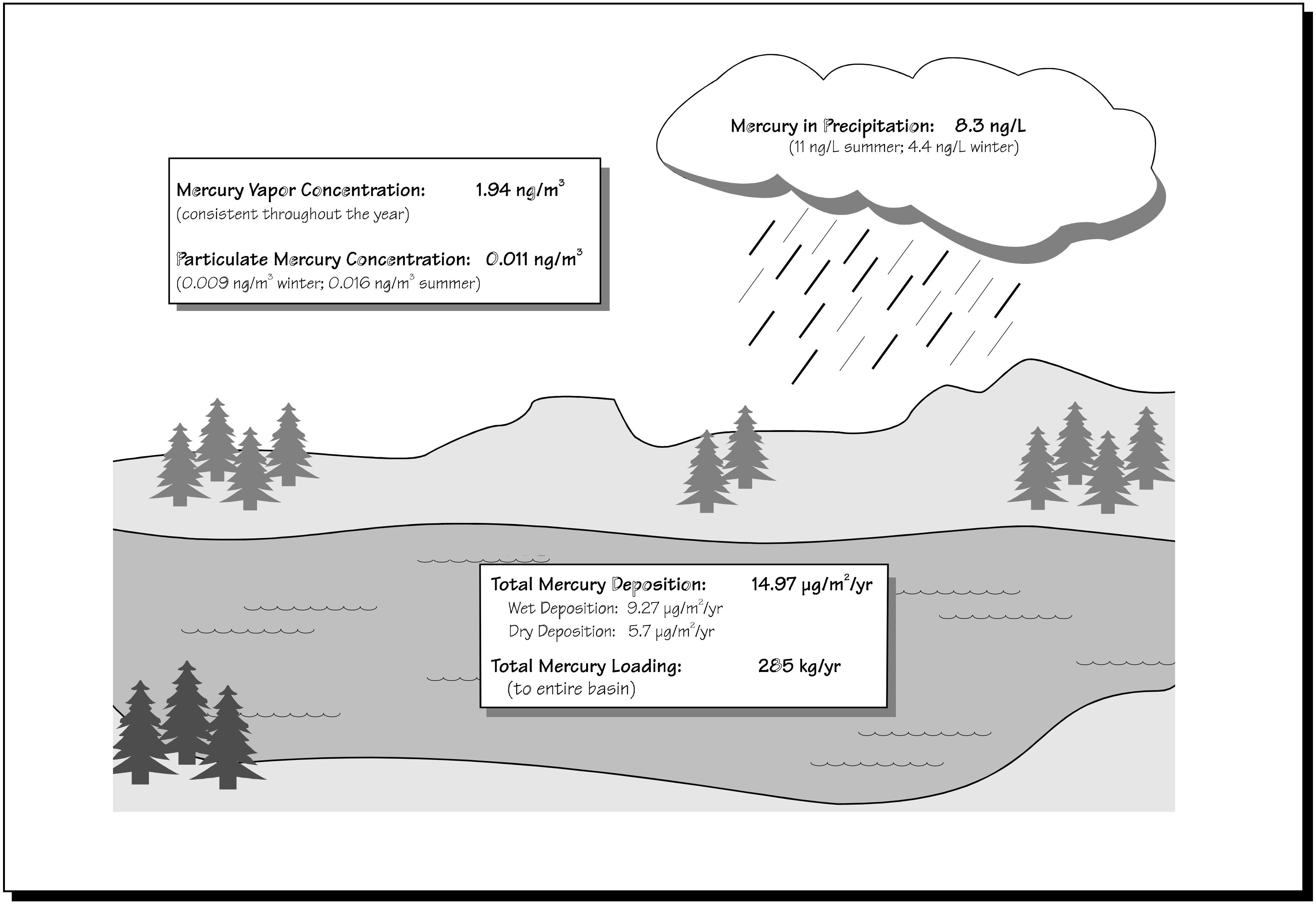
Atmospheric mercury concentration and deposition in Lake Champlain was investigated at a location just east of the Lake in Underhill, Vermont, between 1992 and 1994 (Scherbatskoy et al. 1997). The atmospheric concentration of mercury was measured as gaseous and particulate phases in the ambient air and in precipitation (snow, rain) (see sidebar). Findings on the atmospheric mercury levels near Lake Champlain are presented below and in Figure IV-7:

> **Principal Phases of Atmospheric Mercury**
>
> ***Gaseous (Vapor) Phase Mercury***: consists primarily of elemental mercury (Hg^o; predominant form in the atmosphere), although divalent mercury (Hg^{+2}) may also be present; Hg^{+2} is deposited more rapidly than elemental mercury.
>
> ***Particulate Phase Mercury***: consists of fine particles that are not readily deposited from the atmosphere; bound Hg^{+2} is the predominant form and exists as the nucleus in dust particles.
>
> ***Mercury in Precipitation***: consists of mercury either as dissolved gas or bound to fine particles; primarily particulate Hg^{+2} that has been taken up by rain droplets.

♦ *Atmospheric gaseous mercury concentration.* Average gaseous concentration for 1993 was 1.94 ng mercury/m^3 (comparable to other sites near the Great Lakes), staying relatively constant throughout the year (Table IV-5). Concentrations are typically two orders of magnitude (100 times) higher than particulate mercury.

♦ *Average particulate phase mercury concentration.* Unlike the gaseous phase, particulate phase mercury in the air exhibited seasonal variability, with levels higher in winter than in summer (Figure IV-7). The source of the increased particulate mercury concentration in the winter has not been identified, but preliminary meteorological analysis suggests that a more regional influence is important in the transport of particulate phase mercury in the winter, due to higher average wind speeds and colder temperatures during this season. It also is suggested that colder temperatures in the winter may favor condensation of gas onto particles, increasing mercury particulate concentration in the atmosphere (Scherbatskoy et al. 1997). Furthermore, daily mercury concentrations did not fluctuate significantly, as would be expected for strong local sources.

FIGURE IV-7
Atmospheric Mercury in Lake Champlain Basin[a]



[a] Values are annual average, unless otherwise specified.

Source: Scherbatskoy et al. 1997

TABLE IV-5
Comparison of Mean Total Atmospheric Mercury Concentrations [a]
(Gaseous and Particulate Phases and in Precipitation)

Location	Gaseous Phase (ng/m^3)	Particulate Phase (ng/m^3)	Precipitation (ng/L) (volume-weighted)
Lake Champlain	1.94 ± 0.5	0.011 ± 0.007	8.3 ± 5.2
Northern Michigan[b]	NA	0.011	7.9 ± 7.4
Southern Michigan[b]	NA	0.022	10.2 ± 9.8
Northern Wisconsin[c]	1.57 ± 0.4	0.02 ± 0.02	10.5 ± 4.8
Northern Wisconsin[d]	1.8 ± 0.4	0.01 ± 0.02	6.7 ± 5.6

NA = Not Available
[a] Data are means for varying years and periods; different sampling methods for the studies. Gaseous and particulate phase mercury data represent total dry deposition.
[b] Hoyer et al. 1995; Keeler et al. 1995
[c] Fitzgerald et al. 1991
[d] Lamborg et al. 1995

Adapted from Scherbatskoy et al. (1997).

- *Average mercury concentration in precipitation.* Levels appeared to be typically higher in the summer than the winter. Partial data from 1994 suggest that the pattern observed in 1993 is typical for this area. Based on meteorological data, precipitation events with the highest mercury levels during the year were associated with regional transport from the south or west; transport from the west occurred only during the summer months (Burke et al. 1995). The observed mercury levels are low; studies have reported mercury concentration in precipitation generally less than 100 ng/L in areas not directly influenced by emission sources. The type of mercury in precipitation was not quantitatively determined (Scherbatskoy et al. 1997); however, other studies of precipitation found mercury to be in the inorganic (Hg^{+2}) form, probably as mercuric chloride (Burke et al. 1995), with only 2 to 10 percent in the organic (methyl) form (Bloom and Watras 1989; Driscoll et al. 1994).

- *Yearly deposition of mercury.* Deposition to the entire Lake Champlain Basin, not just to surface water, was estimated from precipitation (wet deposition) and dry deposition data. Total annual mercury deposition was not very different from that observed at other U.S. locations (Table IV-6). Dry deposition was calculated by using the same monthly proportion of gas deposition to gas concentration reported in Lindberg et al. (1992). Deposition of particulate phase mercury was not a significant source of atmospheric mercury loadings when compared to deposition of mercury in precipitation and from the gas phase. The atmospheric mercury particulate concentration in the area (0.011 ng/m^3) was much lower than the gas concentration (1.9 ng/m^3); therefore, the estimate of mercury dry deposition was based solely on gaseous mercury (which may be an underestimate of deposition to some extent). As shown in Figure IV-7, the annual dry deposition is 5.7 μg mercury/m^2 (with levels higher in the summer than the winter). Mercury deposition in precipitation for 1993

TABLE IV-6
Comparison of Annual Mercury Deposition Estimates[a]

Location	Estimated Wet Deposition ($\mu g/m^2$/year)	Estimated Dry Deposition ($\mu g/m^2$/year)	Total Mercury Deposition ($\mu g/m^2$/year)
Lake Champlain basin[b]	9.27	5.7	14.97
Michigan[c]	5.5-13.0	NA	NA
Little Rock Lake, WI[d]	4.5 ± 2 from rain 2.3 ± 0.3 from snow	3.5 ± 3	10.3 ± 3.6
Minnesota and Northern Wisconsin[e]	NA	NA	12.5

NA = not available

[a] Methods and assumptions for estimating these values varied with studies.
[b] Scherbatskoy et al. 1997
[c] Hoyer et al. 1995
[d] Fitzgerald et al. 1991
[e] Engstrom et al. 1994; Swain et al. 1992

was 9.27 $\mu g/m^2$, with deposition higher in the summer (1 $\mu g/m^2$/month) compared to winter (0.2 $\mu g/m^2$/month). The pattern for mercury deposition in precipitation is probably a result of higher mercury concentrations and higher amounts of rain during the summer; however, insufficient data are available to determine the source of the increased mercury concentration in the summer.

Atmospheric mercury can also enter the lake from snowmelt, which adds higher concentrations of mercury to the rivers emptying into Lake Champlain. The total mercury concentration at three river sampling sites increased two- to six-fold during the peak snowmelt event as compared to the mean levels over the course of the year (Scherbatskoy et al. 1997).

Because the Lake Champlain basin has such a large ratio of watershed to lake surface area, it is important to understand the deposition and cycling of atmospheric pollutants by the largest categories of land use, forests and agriculture, prior to their entry into the lake in runoff. Unlike the Great Lakes watershed, which has relatively more lake surface area, Lake Champlain's water surface area makes up only five percent of the basin area. Therefore, it is important to understand mercury cycling through the terrestrial, atmospheric, and aquatic systems of the Lake Champlain Basin, as ongoing studies are beginning to address.

SOME PROGRAMS RELATED TO TOXICS EMISSIONS AND REDUCTION IN LAKE CHAMPLAIN

Some programs by states have been established to address toxics reduction and to identify and control source emissions in the surrounding region. These efforts may have important impact on atmospheric deposition to Lake Champlain basin. Some of these programs are highlighted below:

- The State of New York recently began a program for reducing toxics through a multi-media approach. The Multimedia Program for Pollution Prevention, when fully implemented, will integrate environmental protection programs across all "media," such as air, water, and land, to correct the problem with single media programs.

- The State of Vermont conducts a regulatory program for the sources of about 288 hazardous air contaminants (carcinogens, chronic systemic toxicants, and short-term irritants). Hazardous Ambient Air Standards are established for each of these contaminants, with requirements imposed on new and existing sources (excludes fossil fuel combustion) emitting any of these contaminants in excess of a pre-determined "action level" for each pollutant.

- New York and Vermont established a Permit Exchange Agreement in accordance with the 1988 Memorandum of Understanding, in which both states are informed of permitted projects in the basin. It allows the affected public to participate in the comment and review process for the permits. Potential toxics sources subject to this agreement include air pollution sources within 80 km of each state border that annually emit 50 tons of volatile organic compounds, sulfur dioxides, nitrogen oxides, carbon monoxide, or particulate matter, or 5 tons of lead, and/or are subject to Title V of the CAA.

Addressing Toxic Contamination Reduction in Lake Champlain

The Lake Champlain Basin Program (LCBP) (see sidebar) has been the institutional framework for coordinating the development of a comprehensive pollution prevention, control and restoration plan for the future of Lake Champlain. The final plan was released in October 1996 (LCBP 1996), following public meetings that allowed interested parties to comment on the an earlier plan. It is anticipated that the objectives of the final plan will be implemented by the Lake Champlain Steering Committee, which is represented by environmental officials from New York, Vermont, and Quebec.

One major issue addressed in the plan is the prevention of pollution from toxic substances in order to protect public health and the Lake Champlain ecosystem. Implementation of the plan will require coordination along all levels of government, organizations, and individuals. The plan identified the following major technical and policy issues involved in determining the most appropriate and cost-effective actions to reduce toxic contamination of Lake Champlain.

Lake Champlain Basin Program

The Lake Champlain Special Designation Act, sponsored by senators from Vermont and New York, was signed in 1990 and states that Lake Champlain is a resource of national significance. The intent of the Act is to create a comprehensive plan for protecting the future of Lake Champlain and its watershed. The coordination of the activities stated in the Act is the responsibility of the Lake Champlain Basin Program (LCBP), which is jointly administered by the U.S. EPA, the States of Vermont and New York, and the New England Interstate Water Pollution Control Commission. EPA was given $10 million in funds for five years to develop a comprehensive pollution prevention, control, and restoration plan for Lake Champlain; the final plan was released in 1996. Other cooperating agencies include the U.S. Fish and Wildlife Service, U.S. Department of Agriculture, U.S. Geological Survey, NOAA, and National Park Service. Formal involvement of Quebec is through the Lake Champlain Steering Committee.

- *Define scope of toxics reduction effort.* Research is needed to define the extent of problems related to toxic pollution in tributaries and in the air. Efforts should be made to improve source identification, with attention given to reducing both nonpoint and point sources through all media and remediating current sources of contamination throughout the Basin.

- *Focus efforts on Lake Champlain pollutants of concern and sites of concern.* The List of Toxic Substances of Concern was established by LCBP, which identified mercury and PCBs as Group 1 chemicals and meriting highest priority for management action (see sidebar). These two pollutants are found in the sediment, water, and biota at levels above appropriate standards or guidelines in Lake Champlain. Because of limited resources to study and monitor toxic substances, assessments should focus on specific sites where contamination is known.

Toxic Substances of Concern in Lake Champlain by Priority Group

Group 1: PCBs, mercury

Group 2: Arsenic, cadmium, chromium, dioxins/furans, lead, nickel, PAHs, silver, zinc

Group 3: Ammonia, persistent chlorinated pesticides, phthalates, chlorinated phenols, chlorine, copper

Group 4: Other contaminants known to be used or known to occur in the Basin (e.g., volatile organic compounds such as benzene, pesticides such as atrazine, strong acids and bases)

Source: LCBP 1996.

- *Identify sources and quantify loads of toxic substances.* Efforts to determine sources of toxic substances within Lake Champlain have been initiated, and few "active" sources have been identified. Two major information gaps in this area that need to be addressed include the extent of contamination from outside the basin, and the role of historical sources (e.g., discharged lead batteries released into the waterbody) and contaminated sediment.

- *Adopt strategy to prevent pollution.* Pollution prevention techniques (e.g., source reduction) may be used to achieve reductions at the source of the problem, and eventually reduce pollutants in the lake. Vermont and New York have initiated programs to accomplish this goal, although these programs are not yet integrated into the existing pollution control programs.

- *Establish firm and defensible toxic reduction goals.* The current chemical-by-chemical approach to managing pollution in Lake Champlain cannot account for the impact of cumulative or combined effects, and does not protect against unregulated (and potentially more toxic) chemicals entering the lake; therefore, the reduction strategy should be expanded to address toxic substances that do not yet exceed human health standards or cause measurable impacts within the basin.

In addition, as part of these efforts, relevant information gathered from other programs, such as those initiated in the Great Lakes, should be applied to Lake Champlain. As discussed in Section IV.A, the Great Lakes Water Quality Agreement Parties adopted a long-term goal for virtual elimination of sources of specific pollutants. This effort demonstrates the importance of binational cooperation to address concerns related to reducing toxic emissions in the Great Lakes, as well as in Lake Champlain.

IV.C Chesapeake Bay

The Chesapeake Bay, largest of the 130 estuaries in the United States, was the first in the nation to be targeted for restoration as an integrated watershed, airshed, and ecosystem. The 166,000 km^2 drainage basin (or watershed), shown in Figure IV-8, covers parts of six states (Delaware, Maryland, New York, Pennsylvania, Virginia, and West Virginia) and the District of Columbia, and includes more than 150 rivers and streams. The major tributary basins within the Chesapeake Bay watershed are shown in Figure IV-9, which is found later in this section.

Stretching from Havre de Grace, Maryland, to Norfolk, Virginia, the Chesapeake Bay is 314 km long, and ranges from 5 to 56 km wide. The Bay has over 90,000 km of shoreline (more than the entire West Coast of the continental United States) and a surface area of approximately 30,800 km^2. Generally shaped like a shallow tray, the Bay's average depth, including all tidal tributaries, is only 6 meters, with a few deep troughs running along much of its length that average 18 to 21 meters, and reaching 53 meters at the deepest point. To visualize the relatively large watershed in contrast to the small Bay volume, imagine that the Bay's watershed is reduced to the size of this page; the relative size of the Chesapeake Bay would be a section 7.2 inches by 0.9 inches in the lower right hand corner, and the average depth of the Bay would be represented by one sixtieth the thickness of the paper (see also Figure IV-8).

Economic Highlights of Chesapeake Bay

- In 1992, the dockside value of commercial shellfish and finfish harvests from Chesapeake Bay was close to $80 million.
- In 1993, more than 175,000 pleasure craft (e.g., sail boats) were registered in the Bay.
- Close to 1 million anglers in Maryland and Virginia made an estimated 600,000 fishing trips in 1991. Recreational fishing in these states is estimated at more than $1 billion annually.
- The Chesapeake is a key commercial waterway, and home to two of the nation's five major North Atlantic ports (Port of Baltimore, MD, and Hampton Roads Complex, VA). More than 90 million tons of cargo were shipped via the Bay in 1992.

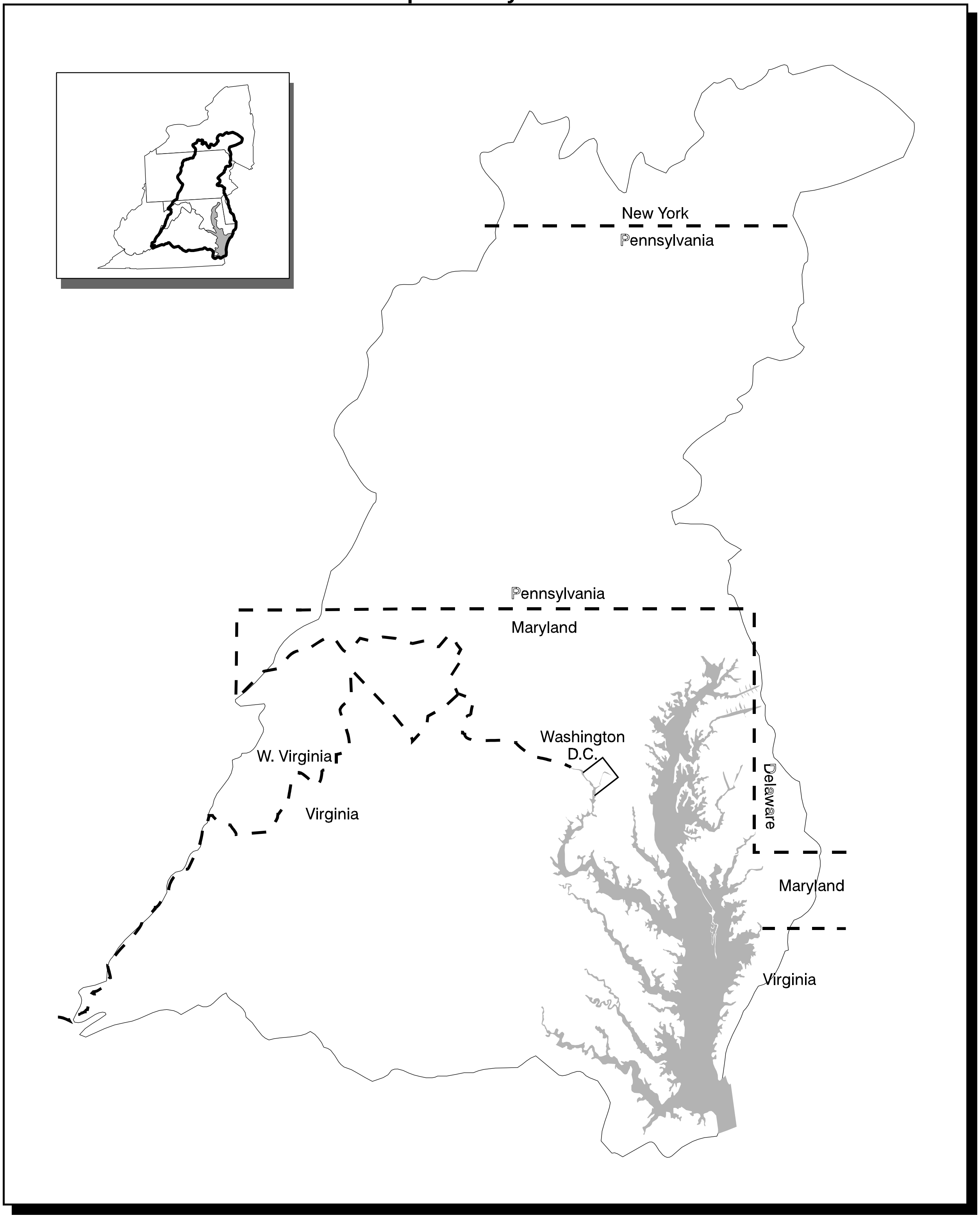
Supporting 295 species of finfish, 45 species of shellfish, and 27,000 plant species, the Chesapeake Bay is a national ecological treasure. The Chesapeake Bay is also home to 29 species of waterfowl and is a major resting ground along the Atlantic Migratory Bird Flyway. Every year, one million waterfowl winter in the Bay's basin. Economic highlights of the Chesapeake Bay are presented in the sidebar above. In all, the Chesapeake is a commercial and recreational resource for more than 14 million Bay basin residents.

The remainder of Section IV.C presents information on: the Chesapeake Bay Program; atmospheric deposition of nitrogen to the Chesapeake Bay; and atmospheric deposition of toxic contaminants to the Chesapeake Bay.

Chesapeake Bay Program

Now in its fourteenth year, the Chesapeake Bay Program is a unique, regional partnership that has directed and coordinated Chesapeake Bay restoration since the signing of the historic 1983 Chesapeake Bay Agreement. The principal partners in the Chesapeake Bay Program include the State of Maryland, the Commonwealths of Virginia and Pennsylvania,

FIGURE IV-8
Chesapeake Bay Watershed



the District of Columbia, the Chesapeake Bay Commission (representing the state legislatures), and EPA on behalf of the federal government.

In 1983, EPA identified an excess of the nutrients nitrogen and phosphorus, and the resulting accelerated eutrophication, as the primary reason for the decline in water quality in Chesapeake Bay (U.S. EPA 1983). Excess nutrients stimulate "blooms" of phytoplankton algae, which then sink to the bottom of the Chesapeake. In the bottom waters, decay of the phytoplankton consumes oxygen, which expands the area of anoxic bottom waters (i.e., "dead waters" largely devoid of oxygen and unable to support life). Blooms of algae also reduce light to submerged aquatic vegetation (SAV), resulting in the loss of an important habitat for juvenile fish and crabs. (A recent assessment of the state of the Bay is presented in the sidebar.)

> The state of the Chesapeake restoration and protection effort was described in the latest *State of the Chesapeake Bay* report (CBP 1995b):
>
> "If the health of the Bay could be likened to that of a hospital patient, the doctor would report that the patient's vital signs, such as living resources, habitat, and water quality, are stabilized and the patient is out of intensive care. Some vital signs, such as striped bass and Bay grasses have improved dramatically, while a few, such as oysters, are in decline. Other vital signs are mixed but stable. Nutrients are being reduced, with phosphorus levels down considerably more than nitrogen levels and dissolved oxygen remains steady. Overall, the patient still suffers stress from an expanding population and changing land use, but it is on the road to recovery. Taken as a whole, the concentrated restoration and management effort begun ten years ago has produced tangible results--a state of the Bay that is better today than when we started..."

Using the watershed as the central focus, the Chesapeake Bay Agreement of 1983 recognized the historical decline of the Bay's living resources and recommended a cooperative approach among the federal and state governments within the watershed to address problems defined by the 1978-1983 Chesapeake Bay Research Program. The one-page agreement committed the signatories to work together to "fully address the extent, complexity, and sources of pollutants entering the Bay." The watershed approach of the state-federal partnership was chosen as the most practical method for implementing restoration efforts on both a local and regional scale.

Building on an expanded understanding of the Bay system and increasing experience with on-the-ground implementation within the cooperative basinwide partnership, a new Chesapeake Bay Agreement was signed in 1987 that set forth a comprehensive array of goals, objectives, and commitments to address living resources, water quality, growth, public information, and governance (Chesapeake Executive Council 1987). The centerpiece of the agreement was a commitment to achieve a 40 percent reduction of nitrogen and phosphorus entering the Bay by the year 2000. This measurable goal added a specific direction to ongoing monitoring, modeling, and nutrient reduction implementation programs. Through the 1987 Bay Agreement, the signatories also committed to "quantify the impacts and identify the sources of atmospheric inputs on the Bay system." This seemingly minor commitment at the time set the stage for a decade-long path to formally address atmospheric deposition as an integral component of basinwide pollution reduction strategies and implementation actions.

Atmospheric Deposition of Nitrogen to Chesapeake Bay

This section presents information on the Chesapeake Bay Nutrient Reduction Strategy, an overview of atmospheric nitrogen loadings to the Bay (from modeling of the airshed, to nitrogen loadings estimates, to modeling of the watershed and estuary), and areas of uncertainty and work underway. Although the Chesapeake Bay Agreement and the baywide Nutrient Reduction Strategy focus on two main nutrients, nitrogen and phosphorus, this section focuses mainly on nitrogen because atmospheric deposition, the focus of this report, is a significant pathway of concern for nitrogen loadings only.

NUTRIENT REDUCTION STRATEGY

The Chesapeake Bay Agreement commits the signatories to reduce the "controllable" nutrient loads by 40 percent by the year 2000. Controllable loads are defined as the baseline year loads minus the loads delivered to the Bay under an all- forested watershed (i.e., a watershed providing only natural, uncontrollable sources of nitrogen) within the Bay Agreement signatory jurisdictions (Linker et al. 1996). In other words, controllable loads are defined as everything over and above the total phosphorus or total nitrogen loads that would have come from an entirely forested watershed in the States of Pennsylvania, Maryland, and Virginia, and the District of Columbia, given existing rates of atmospheric deposition. In this definition, point source loads are considered entirely controllable. In addition, for the Bay Agreement, emissions of nitrogen compounds leading to atmospheric deposition are considered uncontrollable. Nonpoint sources may be controllable or uncontrollable.

Sources of Nitrogen Entering the Bay

Sources of the 170.8 million kilograms of nitrogen delivered annually to the Bay include:

- Point source water discharges (23% or 39.3 million kg), such as sewage treatment plants;
- Atmospheric deposition directly to tidal waters(9% or 15.4 million kg) and indirectly to tidal waters (18% or 30.2 million kg); and
- Other nonpoint sources (50% or 85.9 million kg), such as runoff from agriculture and urban areas.

To measure the goal of reducing controllable nutrient loads by 40 percent, the Chesapeake Bay Program established a 1985 baseline of nutrient loads. The 1985 baseline load was defined using 1985 point source loads and a 1984-1987 average load for nonpoint sources. The Chesapeake Bay Program chose the average load of the 1984-1987 period as the base to be representative of nonpoint source loads for all tributaries, because river flow and associated nonpoint source loads may vary depending on rainfall. Table IV-7 presents the 1985 base load and 40 percent reduction target for the major tributary basins of the Bay, and Figure IV-9 presents the locations of the tributary basins. After the year 2000, the tributary nutrient reduction targets (i.e., the 1985 base load minus the 40 percent reduction target) become nutrient caps that are not to be exceeded at any time in the future even in the face of continued population growth and development of the watershed.

In 1992, the basinwide reduction goal was reevaluated and allocated among the ten major tributary watershed basins. The state jurisdictions, with direct involvement of the public, then developed comprehensive tributary-specific nutrient reduction strategies within the individual watersheds. As part of the 1992 amendments to the Chesapeake Bay Agreement, the signatories committed "to incorporate in the Nutrient Reduction Strategies an air deposition component which builds upon the 1990 Amendments to the federal Clean Air Act and explores additional implementation opportunities to further reduce airborne sources of nitrogen entering Chesapeake Bay and its tributaries" (Chesapeake Executive Council 1992).

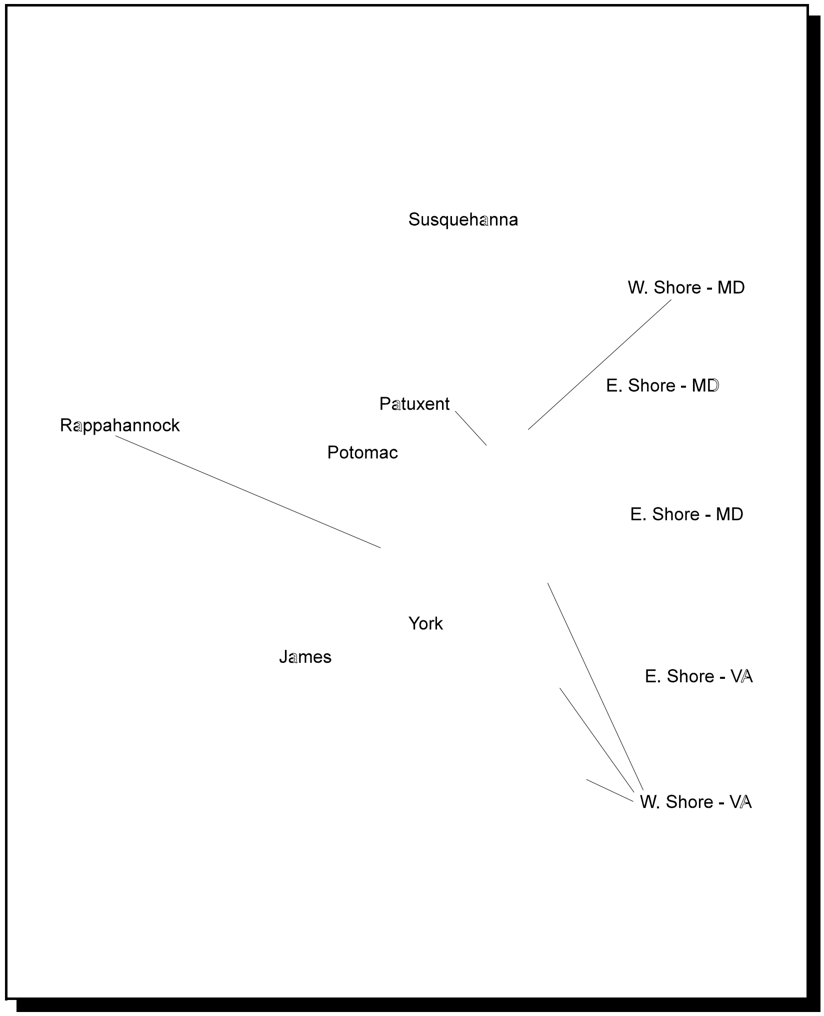
TABLE IV-7
Chesapeake Bay Basin Nutrient Reduction and Loading Caps by Major Tributary Basin
(in millions of kilograms)

Major Tributary Watershed Basin	Nutrient	1985 Base Load	40% Target Reduction	Year 2000 Agreement Loading Cap
Eastern Shore MD	Nitrogen	10.34	2.54	7.80
	Phosphorus	0.82	0.28	0.54
Eastern Shore VA	Nitrogen	0.82	0.18	0.64
	Phosphorus	0.04	0.01	0.03
James[a]	Nitrogen	19.82	6.39	13.43
	Phosphorus	2.80	0.97	1.83
Patuxent	Nitrogen	2.22	0.64	1.59
	Phosphorus	0.24	0.09	0.15
Potomac[b]	Nitrogen	31.16	8.48	22.68
	Phosphorus	2.41	0.78	1.64
Rappahannock	Nitrogen	3.76	1.18	2.59
	Phosphorus	0.39	0.15	0.24
Susquehanna[c]	Nitrogen	52.98	8.30	44.68
	Phosphorus	2.69	1.01	1.69
York	Nitrogen	2.90	0.86	2.04
	Phosphorus	0.42	0.15	0.27
Western Shore MD	Nitrogen	12.02	4.39	7.62
	Phosphorus	0.77	0.30	0.47
Western Shore VA	Nitrogen	1.91	0.54	1.36
	Phosphorus	0.23	0.09	0.14

[a] James loads include only loads from Virginia.
[b] Potomac loads include only loads from Pennsylvania, Maryland, Virginia, and the District of Columbia.
[c] Susquehanna loads include only loads from Pennsylvania and Maryland.

Source: Adapted from CBP 1992.

FIGURE IV-9
Major Tributary Basins of the Chesapeake Bay

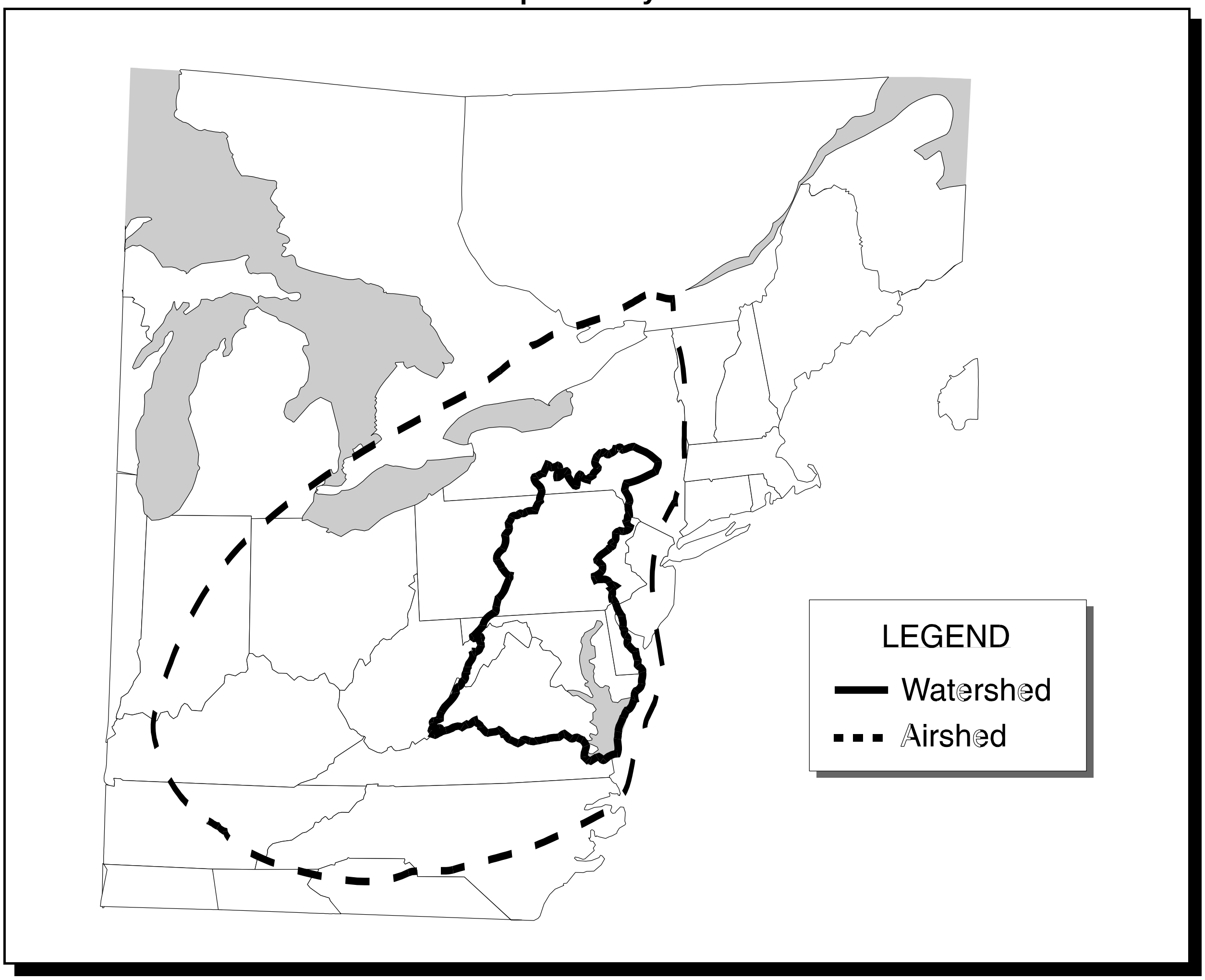


Any reductions in nitrogen loads brought about by programs implementing the CAA are considered to be additional nutrient load reductions separate from the point and nonpoint source reductions identified in the tributary nutrient reduction strategies. CAA implementation is expected to reduce nitrogen loads in Chesapeake Bay beyond the tributary strategy reductions (CBP 1994a). However, these additional reductions may last only a short time; at some time after the year 2000, population growth and increased land development are expected to begin eroding the gains made by the CAA. This expected increase in nutrient loads may make it difficult to meet the caps on nutrient loads to the Bay. Indeed, it was acknowledged at the time the tributary strategies were developed that "achieving a 40 percent nutrient reduction goal, in at least some cases, challenges the limits of current point and nonpoint source control technologies" (Chesapeake Executive Council 1992). To maintain the restoration progress that will be achieved by the year 2000, the technological limits of controls on reductions from point and nonpoint sources may have to be expanded to make further reductions in these areas economically attractive, or other sources of controllable nutrients may have to be considered to achieve cost-effective ecosystem protection in the Bay.

MODELING AIR TO WATERSHED TRANSPORT: THE CHESAPEAKE BAY AIRSHED

A series of linked computer models have been developed by the Chesapeake Bay Program to simulate the transport of nitrogen from its emission sources to the Chesapeake Bay watershed and eventually into the tidal Bay waters. As a first step in establishing the air to tidal waters connection, the "airshed" of the Chesapeake Bay was defined. The boundaries of the airshed were defined as the contiguous areas whose sources "significantly" contributed (i.e., 75 percent) to atmospheric deposition of nitrogen to the Bay and its surrounding watershed (Dennis 1997). These boundaries were delineated by running a series of scenarios on the Regional Acid Deposition Model (RADM), using a predefined point of diminishing return (i.e., when a 50 percent reduction in emissions from large source regions would be expected to produce less than a 10 percent reduction in deposition onto the Bay watershed). The resulting 906,000 km^2 airshed, shown in Figure IV-10, is about 5.5 times larger than the Bay's watershed and includes: all of Maryland, Virginia, Pennsylvania, Delaware, the District of Columbia, West Virginia, and Ohio; most of New York; half of New Jersey, North Carolina, and Kentucky; and parts of Tennessee, South Carolina, Michigan, Ontario, and Quebec (including Lakes Erie and Ontario). (See Chapter III for a description of RADM and Dennis (1997) for more information on the use and limitations of RADM in this study.)

FIGURE IV-10
Chesapeake Bay Airshed

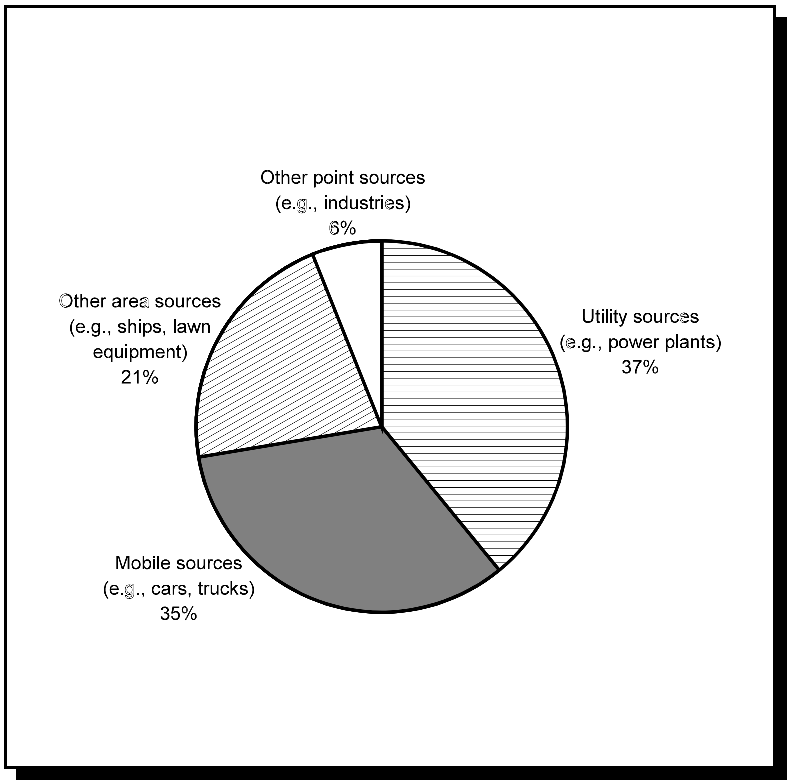


According to the Bay airshed model, about 25 percent of the nitrogen that is deposited on the Bay and its surrounding watershed originates from sources within the Bay watershed. Sources located within the jurisdictions of the Bay Agreement signatories of Maryland, Virginia, Pennsylvania, and the District of Columbia (including those sources that are within the state boundaries but outside of the Bay watershed) contribute about 40 percent of the nitrogen that deposits on the Bay and its watershed (Dennis 1997).

As defined, the Bay airshed, which accounts for 30 percent of all nitrogen emissions in the eastern United States and Canada, accounts for 75 percent of the atmospheric nitrogen deposited onto the Bay and its watershed. The remaining 25 percent of the deposition originates from emission sources outside the defined airshed (Dennis 1997). Therefore, the Chesapeake Bay airshed as defined here is smaller than the actual areas of the United States and Canada that contribute to nitrogen deposition to the Bay watershed. A still unresolved portion of the airshed is the portion that contributes to atmospheric deposition to offshore ocean waters which, in turn, contributes to the influx of nitrogen from coastal waters into the southern Chesapeake Bay (CBP 1994b).

Researchers compared results from the Bay airshed model to emissions inventory data on sources of NO_x emissions and evaluated the contribution of these sources to nitrogen loads to the Bay. As shown in Figure IV-11, data from the emissions inventory indicate that the contributions from utility and mobile sources in the major Bay influencing states (i.e., Maryland, New Jersey, New York, Ohio, Pennsylvania, Virginia, West Virginia) to NO_x emissions are roughly equal and make up the majority of emission sources. Through RADM, these data were confirmed and the patterns of nitrate deposition from the two sources were simulated. The model simulations suggest that utilities contribute a majority of the nitrate that deposits on the western side of the Bay watershed and that nitrate deposition from utility emissions shows a decreasing trend from the western to eastern portion of the watershed (see Figure IV-12[11]). These simulations further suggest that mobile sources, associated with NO_x emissions from the Boston to Washington, D.C., metropolitan areas, contribute a majority of the nitrate that deposits along the Delmarva Peninsula, the Chesapeake Bay itself, and the lower portions of the western shore tidal tributaries (see Figure IV-13). In contrast to utility sources, the simulated deposition from mobile sources shows a decreasing trend from the eastern to western portion of the basin. Model scenarios simulating the effects of a uniform 50 percent reduction in nitrogen emissions from utilities alone and then from mobile sources alone show the same west to east, or east to west, gradients respectively (Dennis 1997).

FIGURE IV-11
NO_x Emission Sources in the Major Bay Influencing States



[11] In Figures IV-12 and IV-13, a rough outline of the watershed and airshed is also shown. Each shaded area in these figures represents the percentage of all emissions that emissions from sources within the shaded area contribute to nitrogen oxides that deposit to the Bay.

FIGURE IV-12
RADM Total (Wet and Dry) Nitrate Deposition from Utility Sources
(as a percent contribution of 1990 Base Case)

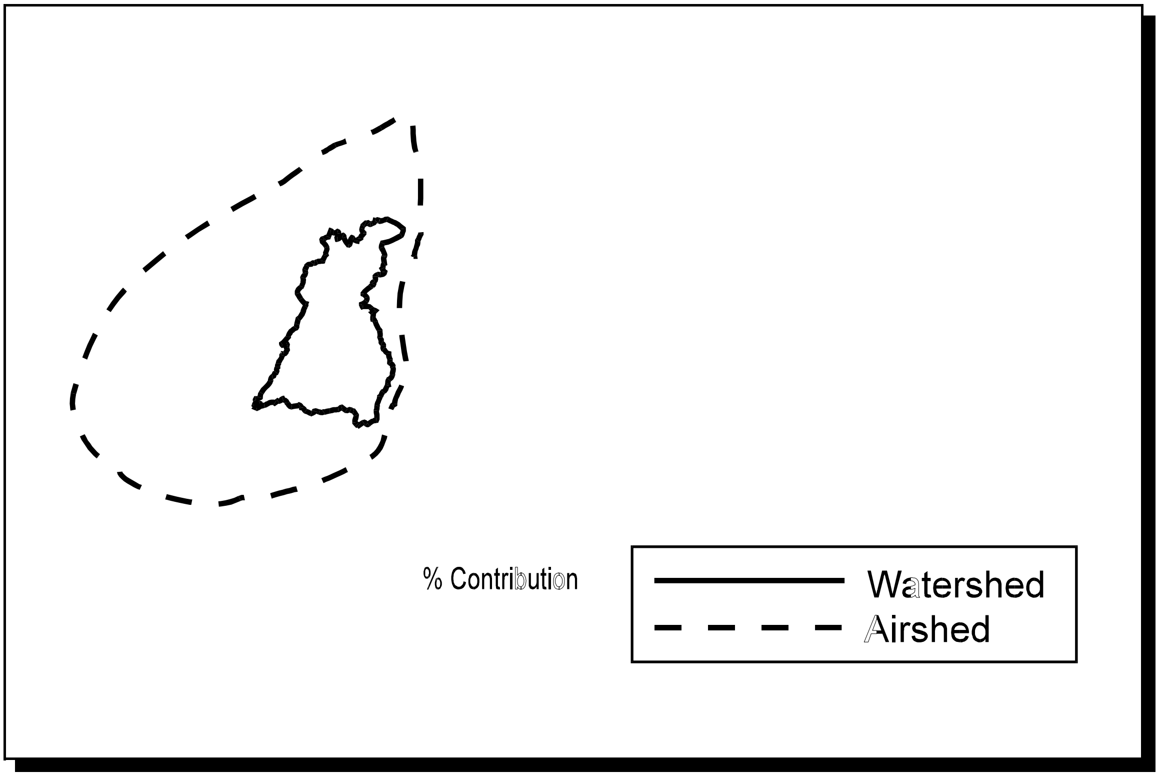
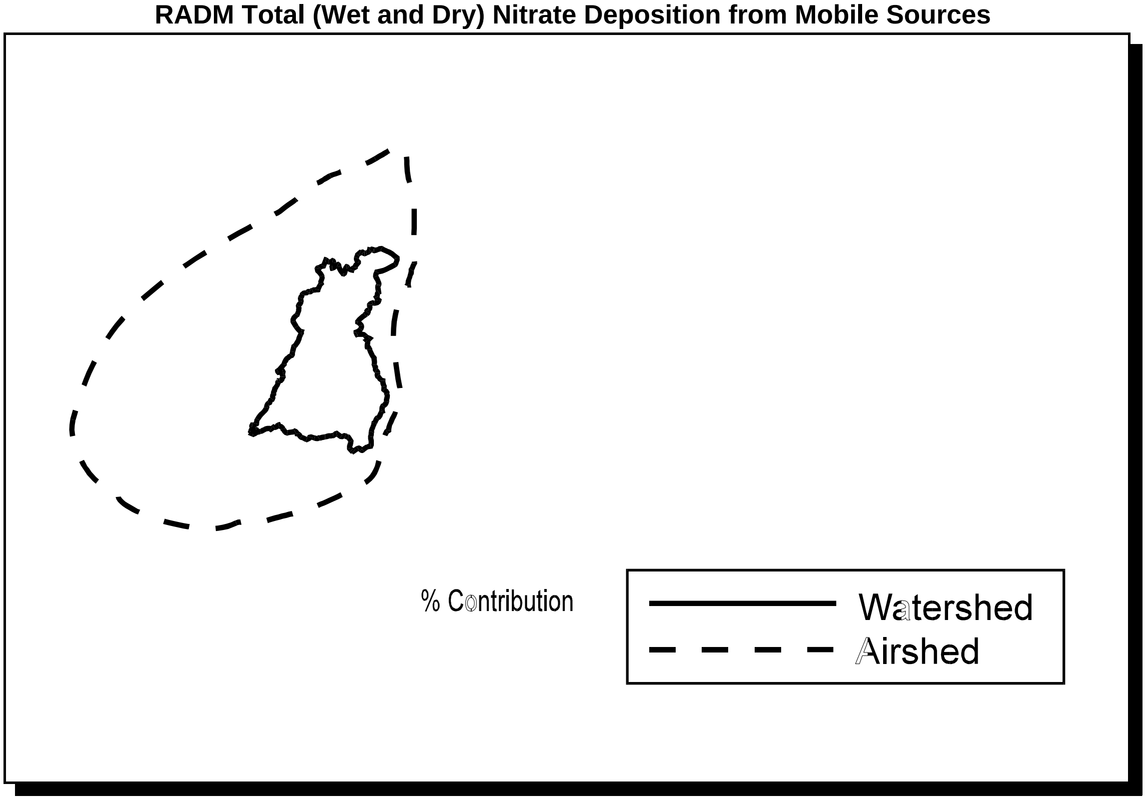


FIGURE IV-13
RADM Total (Wet and Dry) Nitrate Deposition from Mobile Sources



ATMOSPHERIC NITROGEN LOADINGS TO CHESAPEAKE BAY

Atmospheric nitrogen loads from the airshed are transported to the Chesapeake Bay by three routes: direct deposition, both wet and dry, to the Bay tidal waters (i.e., direct loadings); indirect deposition, both wet and dry, to the watershed with subsequent runoff and river transport to the Bay (i.e,. indirect loadings); and deposition, both wet and dry, to adjacent offshore coastal waters with subsequent transport to the Bay through coastal currents. The first two processes, direct and indirect deposition to the Bay, are discussed below, as are some estimates of total loadings to the Bay using both a mass balance approach and computer models. The third pathway, deposition to offshore coastal waters, is the least understood route and is discussed later in this section under areas of uncertainty. Different nitrogen compounds that are measured or estimated in nitrogen loadings are discussed in the box on the next page.

Direct Loadings. The first estimates of atmospheric deposition to the tidal waters of Chesapeake Bay were made through spatial extrapolation of the National Atmospheric Deposition Program (NADP) sites in the Chesapeake watershed (Cerco and Cole 1994). The NADP is a long-term nationwide monitoring program that was started in the 1970s. Based on the annual loads reported by NADP, and an assumption that dry deposition of nitrate is equal to the long-term average of wet deposition of nitrate (Fisher and Oppenheimer 1991; Hinga et al. 1991; Tyler 1988), atmospheric deposition of inorganic nitrogen to the tidal waters of the Chesapeake was estimated as 6.4 million kilograms of nitrate and 1.8 million kilograms of ammonia. In addition, organic nitrogen was estimated as 6.8 million kilograms (Smullen et al. 1982). Studies have explored the idea that atmospheric deposition may contribute a significant proportion of phytoplankton nitrogen demands in coastal areas (Paerl 1985; Paerl 1988; Paerl et al. 1990); phytoplankton require nitrogen, both new and recycled, for growth. Fogel and Paerl (1991), for example, have estimated that 20 to 50 percent of annual new nitrogen demands for phytoplankton in Albemarle-Pamlico Sound, NC, may be supplied by direct atmospheric deposition to the water surface (wet and dry).

Using NADP data, wet deposition directly to the Chesapeake Bay tidal surface waters has been estimated to range from 3.3 to 4.2 million kilograms of nitrate per year (Fisher and Oppenheimer 1991; Hinga et al. 1991; Tyler 1988). Though NADP monitoring data allow initial estimates to be made of atmospheric deposition to the tidal Bay, it is not currently known if the over-land measurements of wet deposition accurately represent over-water wet deposition. To investigate this question, a daily precipitation chemistry site was established on Smith Island, Maryland, in late 1995. This site is providing the first time-series measurements of over-water wet deposition collected on the east coast.

Although the dry deposition to surface water loading rates of nitrogen compounds have been estimated for most nitrogen species over open ocean (Galloway 1985; Duce et al. 1991), these rates may not apply to coastal areas because of the different meteorological processes involved. Through the use of instrumented Chesapeake Bay Observing System (CBOS) buoys owned by the University of Maryland, estimates of nitrogen (HNO_3, NO_2, NH_4) dry deposition rates to the Bay tidal surface waters have been developed (Valigura 1995). These estimates corroborate those given by other investigators to some extent, but still cover a wide range of values, from 0.7 to 2.2 million kilograms of nitrate per year. From this data set, calculations were performed to determine the effect of atmospheric dry deposition on phytoplankton dynamics. This analysis demonstrated that dry deposited nitrogen may provide 10 percent of the annual "new nitrogen" demands by phytoplankton in Chesapeake Bay, and that individual events could supply up to 75 percent of the new demands for periods of several days (Malone 1992; Owens et al. 1992).

Characterization of Nitrogen Compounds

Most atmospheric nitrogen compounds (excluding N_2 and N_2O, which are relatively unreactive in the lower atmosphere) fall into two categories: reactive nitrogen and reduced nitrogen. In addition, some organic nitrogen species arise in the atmosphere from the interaction between nitrogen oxides and certain hydrocarbons. The relative portions of the different nitrogen forms in nitrogen loadings can vary widely based on source types and locations, proximity of sources to receiving waters, atmospheric transport, and physical and chemical transformations. Current estimates are that reactive nitrogen is the largest contributor to atmospheric deposition in coastal waters (40 to 60 percent), with ammonia (20 to 40 percent) and organic nitrogen (0 to 20 percent) also contributing significant amounts.

Reactive nitrogen compounds, primarily oxides of nitrogen, are emitted to the atmosphere through both natural and anthropogenic pathways, overwhelmingly (95 percent) as nitric oxide (NO). Natural NO sources include emissions from soils and generation by lightning; dominant anthropogenic sources include emissions from automobiles, power plants, and biomass burning. The dominant source of reactive nitrogen oxides present in air over North America is high-temperature combustion (e.g., power plants, automobiles). NO generated by combustion reacts quickly in the lower atmosphere, forming nitrogen dioxide (NO_2). The NO_2 is rapidly converted back to NO by ultraviolet light (photochemistry), then NO reacts again, resulting in a cycle driven by volatile organic compounds. From this photochemical cycle, ozone (O_3) is produced. The cycle is broken when NO_2 terminates into stable products, principally nitric acid vapor (HNO_3), and the NO gets used up. The ozone issue is therefore intimately related to the NO_x (defined as NO + NO_2) question. NO_2 slowly deposits to the underlying surface (too slowly to break the cycle), but nitric acid vapor (HNO_3) is easily and quickly deposited. HNO_3 is the source of most of the reactive nitrogen deposited to the earth's surface.

Reduced nitrogen compounds include ammonia (NH_3) and ammonium (NH_4^+). NH_3 is emitted into the atmosphere through both natural and anthropogenic pathways. Natural sources of NH_3 include microbial decomposition of organic nitrogen compounds in soils and ocean waters and volatilization from animal and human wastes. Anthropogenic sources include the manufacture and release of commercial and organic fertilizers during and after application and fossil fuel combustion. Human activities such as manure management and biomass burning exacerbate emissions from otherwise natural processes. NH_3 is a highly reactive compound and has a short residence time in the atmosphere. It is primarily emitted at ground level and quickly deposits to the area near its source unless it reacts with other gaseous chemicals (e.g., SO_2, HNO_3) and is converted to NH_4^+ aerosol (Asman 1994; Langland 1992). NH_4^+ can be transferred regionally as ammonium salts, such as ammonium nitrate NH_4NO_3 and ammonium sulfate $(NH_4)_2SO_4$, and these salts are the primary contributor to NH_4^+ concentrations measured in precipitation. Scavenging of NH_3 by precipitation can also be a major source of NH_4^+ in precipitation.

Organic nitrogen may be a significant fraction of the total nitrogen measured in precipitation (Cornell et al. 1995; Gorzelska et al. 1997; Milne and Zika 1993). Data on the deposition of organic nitrogen has been limited, however, because of the paucity of reliable measurements, the historical variability in analytical techniques and results, and the current lack of suitable and uniform analytical measurement techniques. In fact, only wet deposition of dissolved organic nitrogen (DON) has been addressed. Various estimates for the relative flux of organic versus total nitrogen via wet deposition range from less than 10 percent to greater than 60 percent. The contribution of the unresolved organic fraction may significantly augment the atmospheric deposition of nitrogen to coastal waters. However, in addition to the lack of dry deposition data, there remain many conceptual questions related to source identification and the bioavailability of atmospheric organic nitrogen.

Sources: Luke and Valigura 1997; Paerl et al. 1997; Valigura et al. 1996.

Indirect Loadings. Quantifying indirect loadings of nitrogen to Chesapeake Bay, which refers to the atmospheric deposition of nitrogen to the terrestrial watershed and subsequent transport of the nitrogen from the terrestrial watershed to Bay surface waters, is an important component of the estimate of total atmospheric deposition of nitrogen to the Bay, yet it is largely uncertain. NADP monitoring data provide an initial estimate of the atmospheric deposition to the Chesapeake Bay watershed. Generally, higher deposition levels are found in the northern portions of the basin. In fact, some of the highest readings for atmospheric deposition of nitrate in the NADP monitoring network come from the northern sections of the Chesapeake basin. Greatest uncertainty is in dry deposition of nitrogen, which is not routinely measured by NADP.

The amount of atmospheric nitrogen transferred to surface waters within a given watershed depends on land use, total nitrogen loading from atmospheric, fertilizer, animal waste, and biosolid sources, the amount of soil nitrogen, characteristics of the soil, site rainfall and temperature, the elevation and slope of the land, and the type, age, and health of the vegetative cover. These characteristics vary independently, making it difficult to determine the fate of atmospherically deposited nitrogen over any area of significant size. However, several classification schemes for forested sites have been developed to evaluate a site's potential to retain and leach nitrogen (Melillo et al. 1989; Johnson and Lindberg 1992; Stoddard 1994).

One classification scheme in particular has helped organize thinking about nitrogen retention by classifying forest systems based on stages of nitrogen saturation (i.e., the extent to which the system is saturated with nitrogen; the more saturated a system, the more likely to leach nitrogen) (Stoddard 1994). The classifications range from Stage 0, where forest nitrogen transformations are dominated by plant and microbial assimilation (uptake) with little or no NO_3 export from the watershed during the growing season, to Stage 3, where nitrogen deposition is well in excess of assimilation and has reduced plant and microbial assimilation capacities resulting in greater export of atmospheric nitrogen as well as losses from mineralization of soil organic nitrogen. Study sites in the southern portions of the Chesapeake Bay watershed generally fall into the low nitrate export classification (Stages 0-1), while the northern portions have generally high to medium export classifications (Stages 1-2).

Total Loadings Estimates Using A Mass Balance Approach. The role of atmospheric transport as an important path for nitrogen deposition to estuarine areas was first publicized in 1988 (Fisher et al. 1988). Based on a mass balance analysis using 1984 hydrology data, the authors estimated that one-third of the nitrogen entering the Chesapeake Bay is deposited from the atmosphere. Several subsequent efforts (Fisher and Oppenheimer 1991; Hinga et al. 1991; Tyler 1988) to quantify atmospheric nitrogen loadings to Chesapeake Bay produced "best-estimate loadings" ranging from 25 percent (Fisher and Oppenheimer 1991) to about 33 percent (Hinga et al. 1991) of the total nitrogen loads to the Chesapeake. (A discussion of the uncertainties in a mass balance approach from one of these studies is presented in the sidebar on the next page.)

The approach taken in these mass balance studies can be divided into two components: (1) estimating wet and dry deposition; and (2) estimating nitrogen retention. A central difficulty in mass balance studies is the use of average land use values of nitrogen retention. Nitrogen retention assumptions used in three of the Chesapeake Bay studies are presented in Table IV-8.

TABLE IV-8
Nitrogen Retention Assumptions Used in Chesapeake Bay Loading Studies
(as a percentage of nitrogen loading)

Land Use	Tyler 1988	Fisher and Oppenheimer 1991[a]	Hinga et al. 1991[a]
Forest	95.2-100.0	80.0 (51.0-100.0)	80.0 (25.0-95.0)
Pasture	93.7-99.96	70.0 (51.0-90.0)	80.0 (25.0-95.0)
Cropland	76.0-99.97	70.0	60.0 (45.0-75.0)
Residential	62.0-95.3	35.0 (0.0-70.0)	25.0 (10.0-50.0)

[a] Numbers in parentheses indicate range tested.

Assembling an adequate understanding of long-term behavior when the processes involved are fundamentally episodic is another major challenge of contemporary models. Some studies indicate that the majority of the atmospheric wet deposition occurs during a few episodes (Dana and Slinn 1988; Fowler and Cape 1984), such that the wet-deposited nitrogen (as well as previously dry-deposited nitrogen) is deposited directly to, or flows quickly into, the surface waters without intermediate reduction in concentration. Despite these difficulties, mass balance studies provide a good first-order estimate of nitrogen loading to Chesapeake Bay.

Experimental manipulation at the watershed scale is being conducted at a few U.S. locations (Kahl et al. 1993; Norton et al. 1994). Work from these sites is providing information on the cycling of nitrogen in forested catchments and is fully supportive of the conclusion that atmospheric deposition contributes to nitrogen loading of coastal waters through the export of atmospherically derived nitrogen. Results of these long-term experiments are just beginning to be published. An example is the Bear Brook watershed in Maine. Divided into treatment and control catchments, the treated catchment received increased nitrogen loading in the form of labeled ammonia. The treated watershed response was an immediate increase in stream nitrate export (Norton et al. 1994; Uddameri et al. 1995).

Inherent Uncertainties in Mass Balance Approach

"It would not be difficult to make the [mass balance] calculations appear more elegant by subdividing the watersheds into more land use types, using a detailed data base of land uses, assembling more detailed lists of point source and agricultural inputs, and using some technique for contouring deposition over the watershed. None of these approaches are likely to make better calculations. More precise and reliable estimates of the magnitudes of inputs of atmospherically-deposited nitrogen to coastal waters will require significant advances in the understanding of many processes responsible for the behavior of nitrogen in terrestrial ecosystems and in rivers and streams."

Source: Hinga et al. 1991.

Total Loadings Estimates Using the Chesapeake Watershed and Estuary Models. The Chesapeake Watershed Model (discussed in more detail below) is one approach to disaggregating the separate components of terrestrial and river nitrogen dynamics in the basin, along with the temporal effects of high loading during rainfall events. The estimate of atmospheric nitrogen loads for both direct and indirect deposition is 27 percent of the annual nitrogen load delivered to the Chesapeake Bay. This estimate was developed using the 1992 Watershed Model (Linker et al. 1993) and is consistent with the range reported by Chesapeake Bay mass balance studies (i.e., 25 to 33 percent). Further refinements are being made to the Watershed Model and an update of the estimate of atmospheric deposition is expected by September 1997.

To estimate wet deposition, the Chesapeake Bay Program combined output from a regression model developed from NADP weekly and daily precipitation chemistry measurements with data from the NOAA rainfall network. This approach yields daily estimates of rainfall to 74 sub-basins of the Chesapeake Bay watershed. Dry deposition was assumed to be equal to wet deposition for over-land areas and 44 percent of wet deposition for over-water areas. Indirect atmospheric loadings from the over-land portion of the watershed were estimated using the Chesapeake Bay Program Watershed Model.

The estimate of a 27 percent contribution of atmospheric deposition to total nitrogen loadings to the Chesapeake Bay falls within the range reported for other major eastern and Gulf coast estuaries, which are discussed in Section IV.D and summarized in Table IV-11 in that section.

MODELING THE CHESAPEAKE BAY'S WATERSHED AND ESTUARY

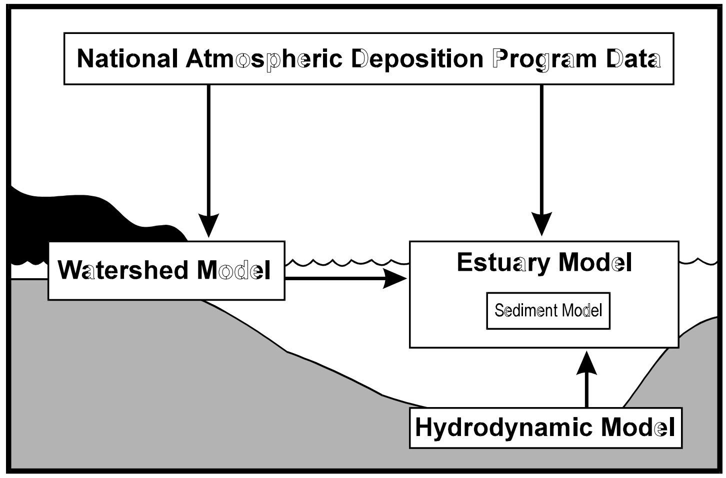
Water quality models of the Chesapeake Bay's watershed and estuary have been in use since the mid-1980s (CBP 1987; Donigian et al. 1991; Hartigan 1983; Thomann et al. 1994). The 1987 Bay Agreement's 40 percent nutrient reduction goal was based, in large part, on findings from these models.

The first model of the Bay watershed was completed in 1982 and provided a basin-by-basin assessment of the relative importance of point source and nonpoint source controls of nutrients (NVPDC 1983). Subsequent refinements of the Watershed Model established the importance of animal waste management in the watershed (Donigian et al. 1991), the delivery to the Bay of atmospheric deposition loads from the watershed (Donigian et al. 1994), and the development of tributary allocation loads of nitrogen and phosphorus to achieve the 40 percent nutrient reduction goal (Thomann et al. 1994).

In a parallel effort, the first model of the Chesapeake estuary was completed in 1987 to evaluate the impact of nutrient reduction scenarios on the Bay's dissolved oxygen concentrations (CBP 1987). Results from the steady-state Estuary Model indicated that a 40 percent reduction in nutrient loads would significantly reduce anoxia (dissolved oxygen concentrations less than 1 mg/L) in the Bay mainstem during average summer (June-September) conditions (CBP 1988). The 40 percent controllable nutrient reduction goal, under the 1987 Bay Agreement, was based in large part on these findings.

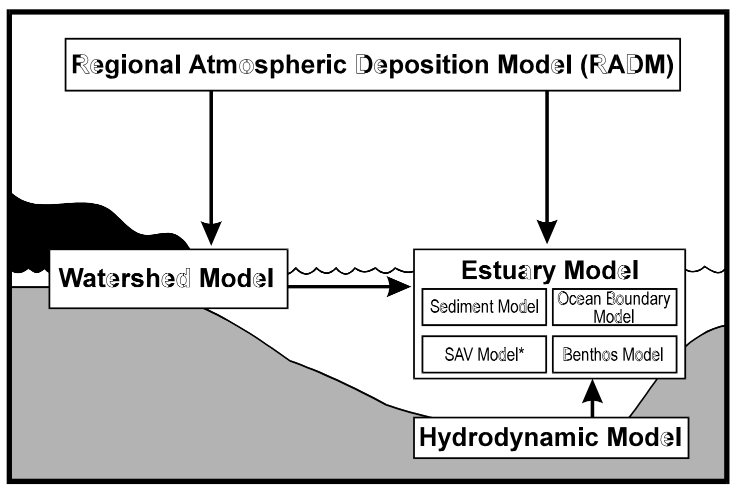
A reevaluation of the Bay's nutrient reduction goal and a review of the progress made in reducing nutrients was scheduled for 1992. In advance of this reevaluation, researchers began refining and integrating the Watershed and Estuary Models (Figure IV-14). For example, the Watershed Model was updated with greater detail of agricultural and atmospheric sources and was linked to the Estuary Model (Donigian et al. 1994). Providing a predictive framework for determining nutrient loads delivered to the tidal Bay under different source reduction scenarios, the Watershed Model simulated delivered nutrient loads with changes in land use practices and levels of wastewater treatment (Thomann et al. 1994). The Estuary Model was upgraded to add a sediment processes model and a hydrodynamic model, and was linked with the Watershed Model to accept Watershed Model nutrient loads as data input (Cerco and Cole 1994; DiToro and Fitzpatrick 1993; Johnson et al. 1991).

FIGURE IV-14
Watershed and Estuary Model Integration



The integrated Watershed and Estuary Model of the Chesapeake Bay was used to estimate water quality improvements that would be realized upon reaching the Bay Agreement goal of reducing controllable nutrients by 40 percent. Through the application of these models, the Bay Program partners established the Bay Agreement tributary nutrient allocations of nitrogen and phosphorus to be achieved by the year 2000 and maintained thereafter.

FIGURE IV-15
Integrated Model Improvements



* SAV = submerged aquatic vegetation

While this initial integrated model (Figure IV-14) could simulate the effects of atmospheric nitrogen deposition on water quality, it could not project the ultimate influence of changes in total nitrogen loadings (i.e., including loadings other than from the air) to the Bay. To provide for this predictive capacity, the Chesapeake Bay Program recently configured the Bay Watershed Model to accept daily atmospheric loadings by land use category (i.e., forest, pasture, cropland, and urban) (Linker and Thomann 1996). The Bay Watershed Model can now simulate the transport of increased or decreased atmospheric loadings to the Bay tidal waters along with nutrients from other land-based point and nonpoint sources. The Estuary Model is being upgraded to simulate basic ecosystem processes of submerged aquatic vegetation (SAV), benthic microorganisms, and major zooplankton groups. In addition, EPA's RADM is being directly linked to the Watershed and Estuary models. This new integrated model, functionally linking the airshed, watershed, estuary, and ecosystem, is expected to be completed in mid-1997. With these refinements, the integrated model (Figure IV-15) will simulate and evaluate the overall loads of controllable and uncontrollable nitrogen from the surrounding airshed and watershed, and the impact of these loads on the ecosystem. This model will be one of the tools used by Chesapeake Bay Program state and federal managers to formulate additional nitrogen reduction steps needed to achieve the 40 percent reduction goal and maintain the cap on nutrient loadings after the year 2000.

WATER QUALITY BENEFITS FROM REDUCING NITROGEN EMISSIONS

Using the Watershed Model, several scenarios were developed to examine the effectiveness of air emission controls compared to traditional point source and nonpoint source controls on the delivery of nutrient loads to the Chesapeake Bay. The Watershed Model scenarios were:

- *Base Case Scenario*: This scenario represents the base year 1985 loads to the Chesapeake Bay.

- *Bay Agreement Scenario*: This scenario represents the 40 percent controllable nutrient load reduction to be achieved by the year 2000 (as discussed under Nutrient Reduction Strategy in this section).

- *Bay Agreement plus CAA Scenario*: The scenario represents the controls on point and nonpoint source loads through the Bay Agreement, plus the atmospheric load reductions expected under full implementation of the CAA titles I (reductions in ground level ozone), II (mobile sources), and IV (utility sources).

- *Bay Agreement plus Ozone Transport Commission (OTC) Scenario*: The scenario evaluates reductions from the controls on point and nonpoint source loads through the Bay Agreement, plus the effects of implementation of CAA titles I, II and IV, as well as additional nitrogen reductions to reduce ground level ozone in the mid-Atlantic and New England metropolitan regions as called for by the Ozone Transport Commission.

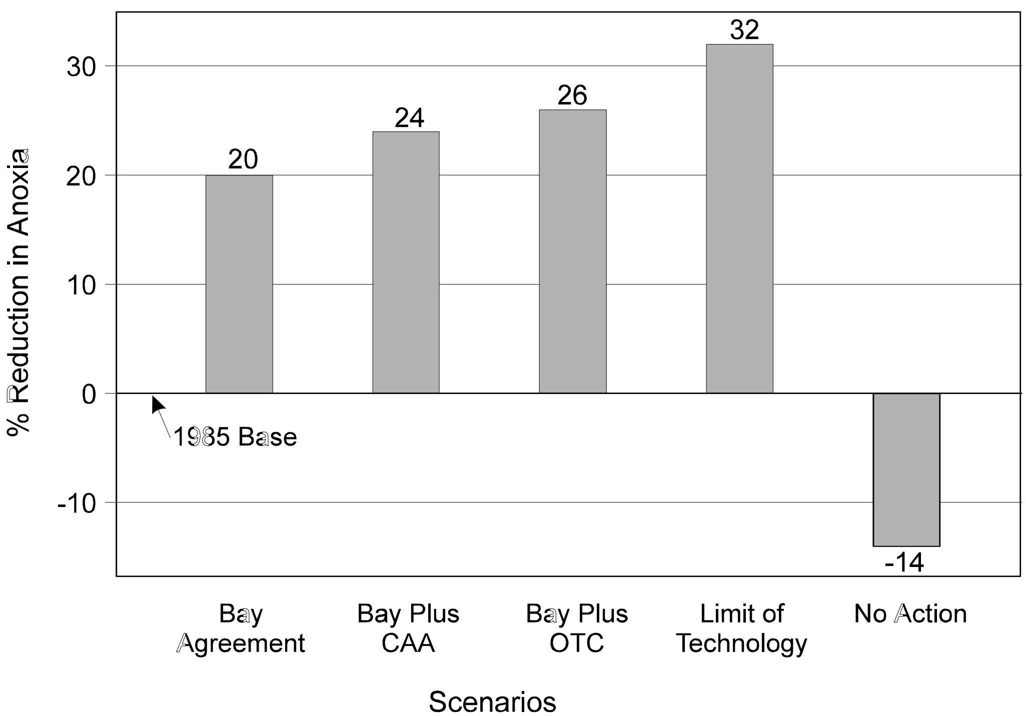
- *Limit of Technology Scenario*: This scenario estimates the nutrient reductions that may be realized using the current practical limit of point and nonpoint source control technologies, including conservation tillage for all cropland implemented; the Conservation Reserve program fully implemented; nutrient management, animal waste controls, and pasture stabilization systems implemented where needed; a 20 percent reduction in urban loads; and point source effluent controlled to a level of 0.075 mg/L total phosphorus and 3.0 mg/L total nitrogen. This scenario is significant because it determines the limit of currently feasible control measures.

- *No Action Scenario*: This scenario represents loads that would occur in the year 2000 given population growth and projected changes in land use. The controls in place in 1985 were applied to the year 2000 point source flows and land use, representing the loading conditions without the nutrient reductions stipulated in the Bay Agreement.

These reduction scenarios are part of an effort to evaluate options for achieving the 40 percent nutrient reduction goal. Land-based nonpoint source and wastewater treatment facility-based point source reduction actions, planned for implementation in many Chesapeake tributary watersheds, are approaching the limits of technology. Options for reductions in air emissions are being explored for maintaining the targeted 60 percent nutrient loadings cap beyond the year 2000 in the face of a growing population and resultant development in the watershed. Different options will have different costs and effectiveness with regard to water quality improvements, and a range of options should be evaluated to find the best approach.

The water quality improvement from the expected reduction in nitrogen emissions under each scenario are shown in Figure IV-16. The improvement in water quality reflects the estimated reductions in Bay bottom waters having no dissolved oxygen (i.e., reduction in Chesapeake anoxia or "dead waters"). Decreased nitrogen loadings will result in decreased water column nitrogen which will, in turn, decrease the growth of algae and improve the level of light penetration necessary to support the critically important SAV (Batiuk et al. 1992; Dennison et al. 1993; Thomann et al. 1994).

FIGURE IV-16
Reductions in Anoxia Under Nutrient Reduction Scenarios



The CAA and OTC scenarios indicate that these controls provide for nitrogen loading reductions and water quality improvements above and beyond those provided by implementation of the Chesapeake Bay Agreement commitment of 40 percent reduction in controllable nitrogen. Relative reductions from controls on sources of atmospheric deposition vary by tributary basin, with the more sensitive tributaries being the Susquehanna and the Potomac. These basins receive the highest deposition loads in the Chesapeake watershed and are among the most responsive to reductions in atmospheric deposition.

Though the differences between scenarios in percent reductions in anoxia might seem small, air emission controls could account for a fifth and a third of the baywide nitrogen load reduction goal through CAA implementation and OTC reductions, respectively. Such reductions could make achieving the 40 percent reduction target more feasible, and make maintaining a cap on any further increases in nutrient loadings beyond 2000 possible. These additional reductions are especially important in the face of increasing population and watershed development that studies predict will increase the significance of atmospheric deposition as a source of nitrogen loadings to the Chesapeake Bay in the coming decades (Fisher et al. 1988; Pechan 1991).

AREAS OF UNCERTAINTY AND WORK UNDERWAY

Nitrogen retention, the relative loadings of ammonia and organic nitrogen, and dry deposition to water surfaces are a few of the remaining areas of significant uncertainty in estimating atmospheric nitrogen loads. Several specific examples of areas of uncertainty are discussed below.

- ***Nitrogen retention*** within watersheds makes a big difference in the proportion of the atmospheric contribution to nitrogen loading to the Bay. Different retention assumptions in mass balance analyses lead to an uncertainty in the estimate of this contribution by more than a factor of two.

- ***Ammonia and organic nitrogen*** contribute a large portion of nitrogen deposition, perhaps more than 25 percent of the total atmospheric nitrogen load. However, it is unknown to what degree their sources are controllable, and they may be changing with time. For example, airborne ammonia emissions from agricultural animal operations could increase.

- Estimates of the relative contribution of ***dry deposition*** to the total atmospheric deposition loadings range from 25 to 63 percent (Duce et al. 1991; Levy and Moxim 1987; Logan 1983; Lovett and Lindberg 1986; Sirois and Barrie 1988; Walchek and Chang 1987). Faced with this wide range of estimates, many investigators choose to set the dry deposition loading equal to the measured wet deposition loading. This assumption is known to be questionable. While site-specific data to refine the estimate are lacking, recent evaluations indicate that dry deposition to tidal water surfaces is about 25 percent of wet deposition to tidal water surfaces (Luke and Valigura 1997).

- ***1990 baseline emission estimates*** continue to be refined. Estimates of emissions from off-road vehicles have been significantly improved. Ship emissions in harbors are suspected to be significantly underestimated (Booz-Allen & Hamilton 1991). While emissions from these sources are not large in the aggregate, they occur close to the Bay tidal surface waters, and thus have an influence greater than their national fractions would imply. Nitrogen emissions from on-road vehicles continue to be a source of uncertainty.

- ***Particulate nitrate*** (which has a low deposition velocity) and ***nitric acid*** (which has a high deposition velocity) are currently indistinguishable by RADM, leading to modeling uncertainty.

- The contribution of atmospheric nitrogen deposition to ***offshore ocean waters*** has not yet been characterized. The ocean waters exchange with waters of the Chesapeake Bay and thus may be a source, or a sink, of nitrogen loads to the Bay.

To reduce existing uncertainties in atmospheric loadings estimates, the Chesapeake Bay Program convened a workshop in June 1994, inviting scientists and managers with expertise and experience in understanding or managing atmospheric deposition. The challenge given to attendees was to construct a list of practical studies that would make the greatest impact on reducing the current uncertainty in estimates of the contribution of atmospheric deposition to declining aquatic ecosystem health. The resulting list (CBP 1995a) is summarized below:

♦ *Priority 1*: Conduct intensive, coordinated, and integrated monitoring studies at special locations within the watershed that characterize wet deposition, dry deposition, and local catchment area.

♦ *Priority 2*: Improve existing atmospheric models (e.g., reduce grid size, account for the effect of mountains).

♦ *Priority 3*: Improve models of chemical retention in watersheds.

♦ *Priority 4*: Improve emissions inventories and projections.

♦ *Priority 5*: Conduct measurements to extend vertical and spatial meteorological and chemical concentration coverage in models.

♦ *Priority 6*: Establish an extensive array of less intensive measurement stations to improve spatial resolution for selected variables.

To improve the cross-media modeling capabilities and to reduce existing sources of uncertainty in atmospheric deposition loadings estimates, the following work is underway through cooperation between EPA, state and federal agencies, and universities:

- Measuring the concentration of atmospheric organic nitrogen within the Chesapeake Bay watershed;

- Measuring dry deposition of nitrate to tidal surface waters of the Bay;

- Investigating the atmospheric deposition of dissolved organic nitrogen and its isotopic composition (delta (δ) ^{15}N);

- Linking daily atmospheric deposition and resultant nitrogen runoff from pasture, forested, and urban lands within the Chesapeake Bay Watershed Model;

- Decreasing the grid size of RADM across the Bay watershed to increase the spatial resolution and improve the resultant model scenario output; and

- Linking RADM with the Watershed, Estuary, and Water Quality models, including simulation of atmospheric deposition to offshore ocean waters and exchange of the ocean waters with Chesapeake Bay waters.

The result of this and other work will become part of the integrated model of the Bay's airshed, watershed, estuary, and ecosystem (discussed above), which is expected to be completed in early

1997. A series of management scenarios, similar to the modeling scenarios discussed above, are also expected to be completed in 1997 to examine the most feasible and cost-effective combination of point source, nonpoint source, and air deposition reductions to meet the Chesapeake Bay Agreement commitment to cap nutrient loadings in 2000 at 60 percent of 1985 controllable base loadings and to ultimately restore the water quality conditions necessary to fully support the Bay's invaluable living resources.

Toxic Contaminant Deposition to the Chesapeake Bay

CHESAPEAKE BAY BASINWIDE TOXICS STRATEGY

The 1987 Chesapeake Bay Agreement committed the signatories to "develop, adopt, and begin implementation of a basinwide strategy to achieve a reduction of toxics consistent with the Clean Water Act of 1987, which will ensure protection of human health and living resources" (Chesapeake Executive Council 1987). The resultant strategy, adopted in 1989, initiated a multi-jurisdictional effort to more accurately define the nature, extent, and magnitude of Chesapeake Bay chemical contaminant problems and to initiate specific chemical contaminant reduction and prevention actions (Chesapeake Executive Council 1989). Building on a two-year reevaluation of the strategy and increased understanding of the nature of the Bay's toxics problems, a revised, farther-reaching strategy was adopted in 1994. The 1994 *Chesapeake Bay Basinwide Toxics Reduction and Prevention Strategy* recognized the contribution of atmospheric deposition as a significant source of chemical contaminant loadings to the Bay. ***Within the basinwide strategy, the signatories committed to establishing a more comprehensive loadings baseline and setting an atmospheric deposition loading reduction target to be achieved over the next decade*** (Chesapeake Executive Council 1994).

In 1991, the Chesapeake Bay Program adopted its first Chesapeake Bay Toxics of Concern list, principally to identify and provide concise documentation on chemical contaminants that adversely affect the Bay or have a reasonable potential to do so. This list provided Chesapeake Bay region resource managers and regulators with a baywide consensus of priority chemicals and the information necessary to target these chemical contaminants for strengthened regulatory and prevention actions or additional research, monitoring, and assessment. Based on ambient concentrations of chemical contaminants and aquatic toxicity data, the toxic pollutants that represented immediate or potential threats to the Chesapeake Bay system were identified. The Toxics of Concern list (see sidebar) includes several pollutants of concern for atmospheric deposition to the Great Waters (cadmium, chlordane, lead, mercury, PAHs, and PCBs). In addition, a Chemicals of Potential Concern list was identified for the Chesapeake Bay (see sidebar) and includes two pollutants of concern for the Great Waters (dieldrin and toxaphene). Clear evidence is lacking that the contaminants on the Chesapeake Bay list of Chemicals of Potential Concern actually cause or have reasonable potential to cause adverse effects in the environment, but the Chesapeake Bay Program believes these

Chesapeake Bay Toxics of Concern

Toxics of Concern: atrazine, benz(a)anthracene,[a] benzo(a)pyrene,[a] cadmium, chlordane, chromium, chrysene,[a] copper, fluoranthene,[a] lead, mercury, napthalene,[a] polychlorinated biphenyls (PCBs), and tributyltin.

Chemicals of Potential Concern: alachlor, aldrin, arsenic, dieldrin, fenvalerate, metolachlor, permethrin, toxaphene, and zinc.

[a] A polycyclic aromatic hydrocarbon (PAH).

chemicals warrant enough concern to be carefully monitored and tracked. For example, a number of the chemicals listed as being a potential concern are either banned or restricted pesticides that have residues still remaining in the ecosystem at elevated levels but below thresholds of concern; others are chemicals of increasing concern due to use patterns or potential for toxicity to Bay resources.

In response to a commitment within the 1994 Basinwide Toxics Strategy, the Toxics of Concern list is currently being evaluated and revised using a risk-based chemical ranking system incorporating source, fate, and exposure/effects ranking factors. In-depth analyses of the top-ranked chemicals will lead to the selection of a revised Toxics of Concern list in 1997.

CHESAPEAKE BAY TOXIC CONTAMINANT ATMOSPHERIC DEPOSITION STUDIES

Studies conducted in the southern Chesapeake Bay in the early 1980s suggest that the atmosphere is a significant source of organic contaminants to the Bay, such as organic carbon (Velinsky et al. 1986) and anthropogenic hydrocarbons, including PAHs (Webber 1993). While these earlier studies demonstrated the potential importance of the atmosphere in supplying contaminants to the Chesapeake Bay, they were limited by their methodologies (i.e., bulk deposition sampling, which is imprecise) and their relatively limited temporal and spatial scope. An assessment of the extent of toxic contamination in the Bay is presented in the sidebar.

To further explore the issue of atmospheric loadings of toxic contaminants to the Bay, the Chesapeake Bay Atmospheric Deposition Study (CBADS) network was established by a team of scientists from the University of Maryland, Virginia Institute of Marine Sciences, University of Delaware, and Old Dominion University. The primary objective of the CBADS network was to provide the best estimate of total annual atmospheric loadings of a variety of trace elements and organic contaminants directly to the surface waters of the Chesapeake Bay. Because accurate estimates of baywide annual loadings require characterizing the spatial and temporal variability in contaminant loads in the atmosphere and in depositional fluxes to the Bay, CBADS collected data to help characterize this variability.

Extent of Toxic Contamination in Chesapeake Bay

Prior to adoption of the 1994 *Chesapeake Bay Basinwide Toxics Reduction and Prevention Strategy*, the Chesapeake Bay Program conducted a two-year, in-depth evaluation of the nature, extent, and magnitude of toxic contaminant-related problems within the Chesapeake Bay. Through the evaluation, no evidence was found of severe baywide impacts from chemical contamination, unlike other problems facing the Chesapeake Bay, such as the impacts from excess nutrients. The Program did document severe, *localized* toxicity problems, adverse effects from chemical contamination on aquatic organisms in areas previously considered unaffected, and widespread low levels of chemical contamination in all Bay habitats sampled.

Existing state and federal regulatory and management programs continue to reduce the input of potentially toxic chemicals to the Chesapeake Bay. Measured concentrations of many of these chemical contaminants in the Bay's bottom sediments, shellfish, fish, and wildlife have also generally declined, although elevated levels occur in several industrialized areas and some increasing trends have been observed. Progress in reducing the point sources of these chemical contaminants is offset by significant nonpoint source inputs of chemical contaminants (e.g., urban stormwater runoff, atmospheric deposition) from increasing development and urbanization of the Bay watershed.

Based on previous studies of wet deposition in the region (e.g., Tyler 1988) and given the resources available for the network, three non-urban shoreline locations -- Wye Institute and Elms Institute, Maryland, and Haven Beach, Virginia -- were selected and sampled from 1990 to 1993. These three sampling site locations, as well as other monitoring sites around Chesapeake Bay, are shown in Figure IV-17. In establishing this initial network, the influence of urban areas was purposely avoided by locating the sites more than 50 kilometers from metropolitan areas (similar to the initial non-urban stations for the Great Lakes deposition network, the Integrated Atmospheric Deposition Network (IADN)). By minimizing possible urban influences, the resulting CBADS loading estimates are considered to be minimum values.

CBADS evaluated atmospheric loadings directly to the Bay only. Although it is most likely that some fraction of the toxic contaminants deposited from the atmosphere to the watershed of the Chesapeake Bay are ultimately transported to the surface waters, this study did not attempt to characterize indirect loadings for two main reasons:

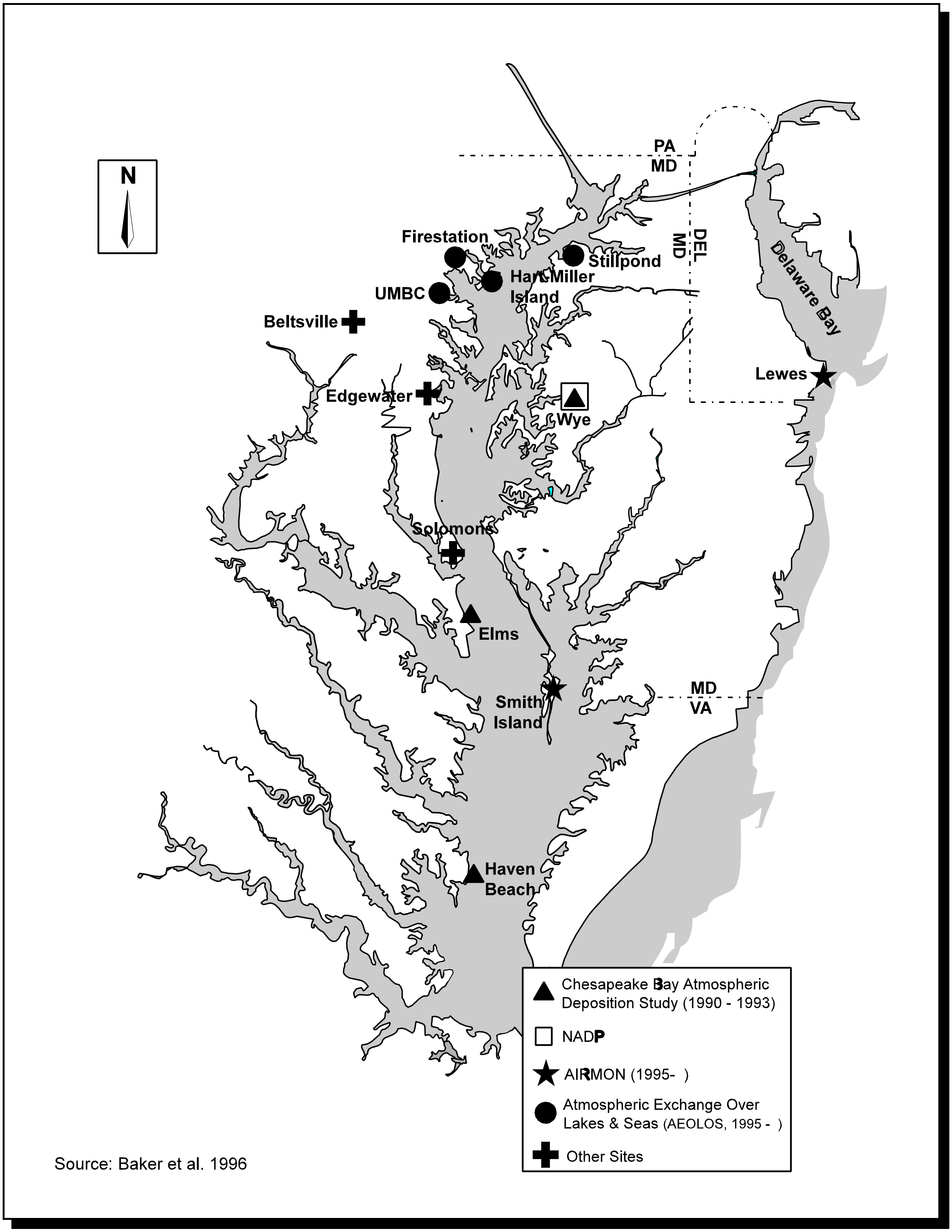
- Because deposition to the various land surfaces is likely much different than that to the water surface, fluxes measured at the shore-based stations cannot be extrapolated with confidence to the watershed; and

- The large uncertainty in the understanding of the fate of materials deposited to the land surface (i.e., the fraction transmitted to the receiving water) precludes the simple estimation of the indirect atmospheric loading of contaminants to the Chesapeake Bay in more than a rough estimate.

The CBADS data on concentrations in air, concentrations in precipitation, and wet and dry aerosol depositional fluxes are presented in Baker et al. (1997) and are summarized below. These data were collected for two groups of contaminants: trace metals (aluminum, arsenic, cadmium, chromium, copper, iron, manganese, nickel, lead, selenium, and zinc); and organic contaminants (14 different PAHs and total PCBs). Cadmium, lead, PAHs, and PCBs are pollutants of concern for atmospheric deposition to the Great Waters. Mercury data are being collected but have not yet been compiled, and data on other Great Waters pollutants of concern were not collected. For detailed results, a discussion of sampling methods, or a description of the limitations of the study, refer to Baker et al. (1997).

Concentrations in Air. Air concentrations for trace metals were determined by measuring the elemental composition of aerosol particles less than 10 μm in diameter. The elemental composition was dominated by the crustal elements aluminum and iron, as well as sulfur (in the form of sulfate). During 1991 and 1992, concentrations averaged over the three sampling sites were 116, 111, and 2,123 ng/m^3 for aluminum, iron, and sulfur, respectively. Trace element concentrations averaged over the same period ranged from 0.16 ng/m^3 for cadmium to 12.6 ng/m^3 for zinc, with lead averaging 3.88 ng/m^3. The fraction of each element derived from non-crustal (e.g., combustion) sources was estimated based on the average concentration of elements in the Earth's crust (Turekian and Wedepohl 1961), and assuming all of the measured aluminum associated with aerosol particles is derived from erosion of soils. In the Chesapeake Bay region, non-crustal sources supply greater than 95 percent of most of the elements measured on aerosol particles (Wu et al. 1994). Arsenic, cadmium, lead, sulfur, and selenium are almost exclusively non-crustal, and are likely introduced into the atmosphere as a result of combustion of fossil fuels and incineration of municipal wastes.

FIGURE IV-17
Sampling Locations for
Chesapeake Bay Toxic Contaminant Atmospheric Deposition Studies



Source: Baker et al. 1996

The measured concentrations of trace elements were generally within a factor of two among the three sampling sites. On an average annual basis, concentrations slightly decrease from north (Wye) to south (Haven Beach), except sulfur, which is 15 percent higher at Elms (1991 and 1992) and Haven Beach (1992) than at the Wye site. The general decreasing trends observed from north to south, along with increasing sulfate, may indicate higher levels of low sulfur combustion sources (e.g., incinerators, vehicles) in the northern reaches of the Chesapeake Bay. In general, the spatial variability in the atmospheric concentrations of trace elements between sites (from north to south) is substantially lower than corresponding temporal trends.

Semivolatile organic chemicals exist in the atmosphere as gases and also are associated with aerosol particles (Pankow 1987). In this study, baywide annual average concentrations of PAHs in air ranged from 16 ng/m^3 for dibenz[*a,h*]anthracene to 2,590 ng/m^3 for phenanthrene. Atmospheric concentrations were quite variable with individual measurements ranging from one-tenth to ten times the annual average concentrations. These variations likely result from sampling air masses coming from differing directions, from changes in local and regional emissions, and from differences in atmospheric degradation and deposition rates. These variations show a seasonal pattern. For example, increased concentrations of gas-phase PAHs, such as pyrene, during the summer months may reflect both higher temperatures (i.e., enhanced volatilization) and increased coal and oil combustion to meet the electricity demand of air conditioning. Increases in the atmosphere of particulate PAHs, such as benzo[*a*]pyrene, may result from local burning of yard wastes and of wood for home heating. Some variation in atmospheric levels of organic chemicals may result from the efficient removal of particulate PAHs by precipitation (Poster and Baker 1996a, 1996b). In general, the magnitude of fluctuations in atmospheric levels of organic chemicals is larger than the corresponding variations in trace elements and sulfur discussed above, suggesting the importance of local combustion sources. Air concentration data were not available for PCBs.

Concentrations in Precipitation. The overall volume-weighted mean concentrations of trace elements in precipitation collected at the three sites range from 0.03 μg/L for cadmium at Elms to 14.6 μg/L for iron at Haven Beach. For lead, the range of overall volume-weighted mean concentrations was between 0.42 and 0.52 μg/L at the three sites. The relative proportion of trace elements in precipitation is nearly identical to that in the Chesapeake Bay aerosol particles, confirming that aerosol scavenging is the source of trace metals to wet deposition. Trace metal wet depositional fluxes are highly variable, changing more than ten-fold between consecutive months with little apparent seasonal dependence. This variability, which was similar at each of the three sites, results not only from fluctuations in the atmospheric inventories of trace metals, but also from changes in the amount of precipitation. On an annual basis, the volume-weighted mean concentrations of most trace metals did not systematically vary between the summer of 1990 and fall of 1993, again suggesting that these rural sites were most strongly influenced by the same regional background signal throughout the study.

While individual precipitation events result in spikes in trace metal deposition at one site that are not observed at the other two locations, these isolated events average out over the year. Annual volume-weighted mean concentrations of trace metals in precipitation are generally within a factor of two among the three sites, with no clear systematic spatial trend observed for all metals.

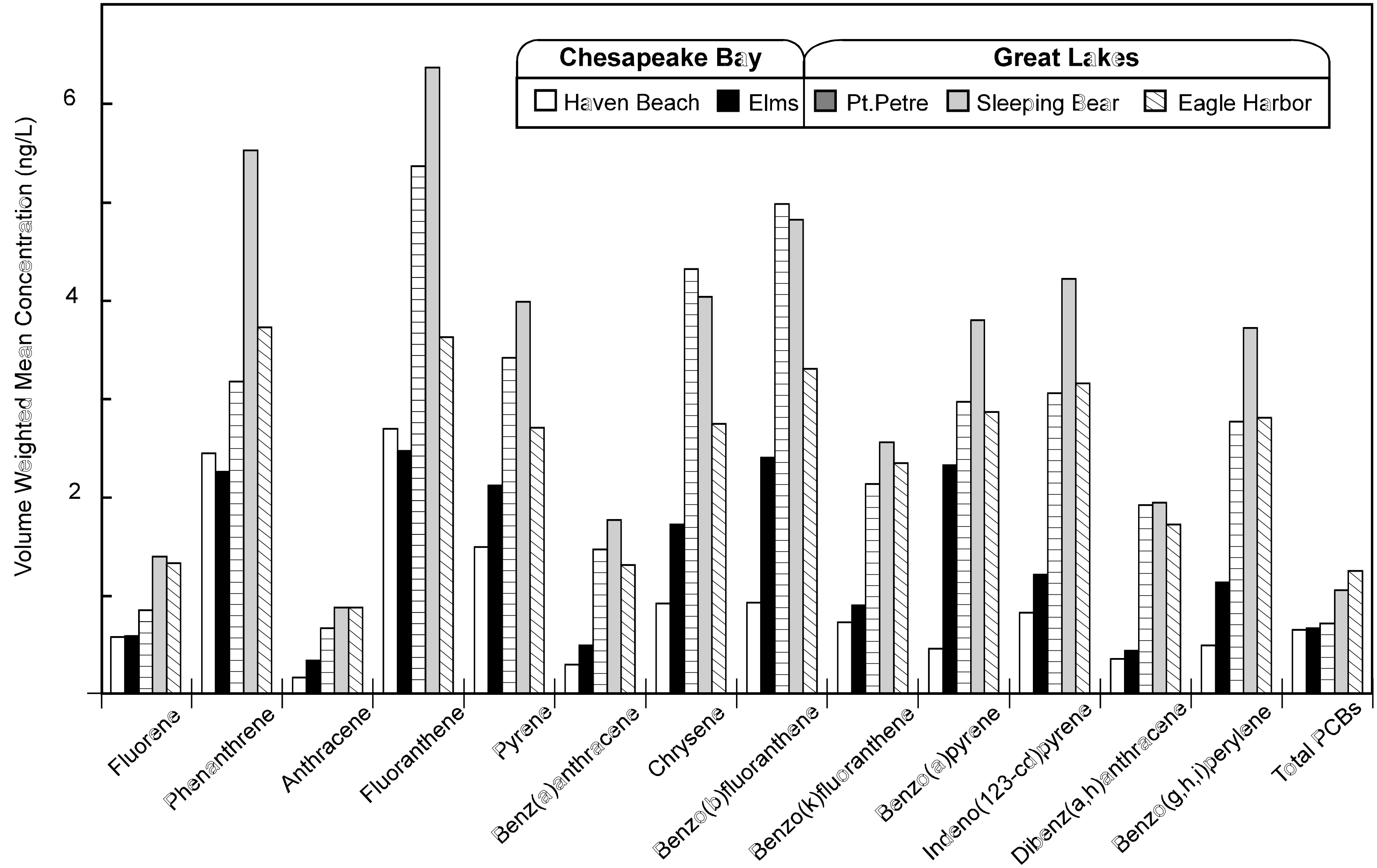
Overall volume-weighted mean concentrations of PAHs in precipitation ranged from 0.3 ng/L for anthracene and dibenz[*a,h*]anthracene at the Havens Beach site to 9 ng/L for pyrene at the Elms site. Volume-weighted mean concentrations for total PCBs were 1.1 and 0.9 ng/L at the Elms and Haven Beach sites (collected from June 1990 through September 1993), and 0.35 ng/L at the Wye site (collected from January to September 1993). As was observed for trace metals, wet depositional fluxes of organic contaminants varied considerably with time, and were dominated by episodic spikes at each location. Extremely high concentrations of some analytes, including pyrene, that were measured in both air and precipitation at the Elms site in the summer of 1990, may have resulted from local vegetation burning. Unlike PAH levels in the atmosphere, concentrations in precipitation did not systematically vary with season. The enrichment at the Elms site is especially pronounced for higher molecular weight PAHs, suggesting a local combustion source (e.g., wood burning for residential heating).

Concentrations of organic contaminants in precipitation measured in this study are consistently lower than those observed in the Great Lakes region (see Figure IV-18). For example, PAH concentrations in precipitation at the Chesapeake Bay sites are one-third to one-half as high as at the three IADN sites located at rural, shoreline sites on Lakes Ontario, Michigan, and Superior (Gatz et al. 1994). In contrast, levels of the same PAHs in the air over Chesapeake Bay are equal to or perhaps higher than those measured over the Great Lakes. Whether the apparent enrichment in PAHs in Great Lakes precipitation relative to that in the Chesapeake Bay region, as shown in Figure IV-18, is due to more efficient scavenging by precipitation in the colder, relatively drier Great Lakes region, or simply reflects methodological differences between the two networks, is unclear.

Wet and Dry Aerosol Depositional Fluxes. Using CBADS data, researchers calculated depositional fluxes (see Baker et al. (1997) for methodologies). Because these "depositional fluxes" are actually gross fluxes directly to the water surface and do not account for net gas exchange across the air-water interface, the term "deposition rates" is used in the remainder of this discussion in place of "depositional fluxes" to be consistent with the rest of the report in the use of the term "flux."

Total annual deposition rates in 1992 ranged from 0.07 mg/m^2 for cadmium at the Wye site to 121 mg/m^2 for aluminum at the Elms site; the highest annual deposition rate for lead was 1.34 mg/m^2 at the Wye site. Not surprisingly, dry aerosol deposition comprises the majority of the total deposition rate for the soil elements aluminum and iron, which occur on coarse particles. Wet deposition contributes between one-third and one-half of the total depositional rate of the remaining trace elements. Naturally, spatial trends in total deposition result from variation in precipitation chemistry and amount, and the trace element inventories associated with aerosol particles (given the considerable uncertainty in the dry aerosol deposition calculation, the same deposition velocity was used at each site). Although a distinct north to south trend in precipitation amount occurred in 1992 (100, 107, and 122 cm, respectively), total annual deposition rates were remarkably similar among the three stations. Total deposition rates were also very similar between years, again indicating that the relatively rare spikes in concentration are dampened against the chronic regional background signal.

FIGURE IV-18
Comparison of 13 PAHs and Total PCBs in Precipitation (1992) from Chesapeake Bay and Great Lakes Sampling Sites



Source: Baker et al. 1996 (Chesapeake Bay) and Gatz et al. 1994 (Great Lakes).

For PAHs, total annual deposition rates for 1992 range from 0.2 $\mu g/m^2$ for anthracene at the Wye site to 10.8 $\mu g/m^2$ for benzo[*b*]fluorathene at the Elms site.[12] Both wet deposition and dry aerosol deposition contribute to total PAH deposition, with dry aerosol deposition becoming relatively more important for the higher molecular weight, less volatile compounds. Total deposition rates for PAHs decrease with time during this study, with the lowest rates occurring during the first nine months of 1993. While some of this decrease is attributed to beginning with anomalously high measurement in the summer of 1990, decreases in both wet and dry aerosol deposition rates continued between 1992 and 1993. The total annual deposition rate for total PCBs is about 3.5 $\mu g/m^2$, with approximately equal contribution from wet and dry aerosol deposition.[13]

Overall, total annual deposition rates for PAHs and PCBs are generally within 50 percent among the sites. Given the uncertainty in the dry aerosol deposition estimates, this percentage indicates little spatial variability when integrating over annual cycles. However, this study did not specifically address the possible influences of urban areas, such as the cities of Baltimore, Washington, and Norfolk, on atmospheric deposition, which may be important.

To place the atmospheric deposition rates calculated in this study in perspective, they are compared to similar estimates made for the Great Lakes region (Figure IV-19). Wet deposition rates for lead and arsenic are almost three times higher in the Great Lakes than in Chesapeake Bay, despite significantly less rainfall (80 versus 110 cm/year); wet deposition rates for cadmium are similar for both regions. Wet deposition rates for PAHs and total PCBs are fairly similar between the two regions, as higher concentrations in Great Lakes precipitation (see Figure IV-18) are offset by lower precipitation amounts. Dry aerosol deposition rate estimates are higher in the Chesapeake region, especially for organic contaminants.[14] In addition, measured aerosol-bound organic concentrations were generally higher than the values used in the Great Lakes dry aerosol deposition calculations (Eisenreich and Strachan 1992). Despite the differences, estimated atmospheric deposition rates are generally within a factor of two between the Chesapeake Bay and the Great Lakes regions, which, given the numerous opportunities for error in these measurements and calculations, is quite good agreement.

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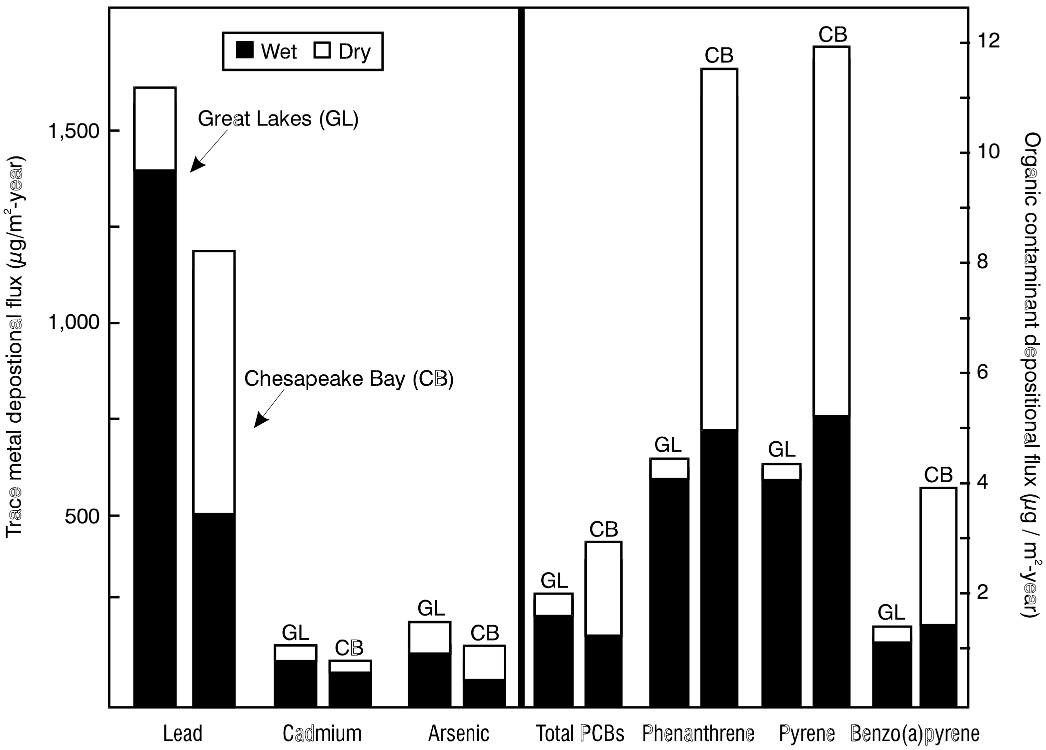
[12] Because the "fluxes" in this study are actually gross fluxes directly to the water surface, these data do not take into account exchange of gaseous organic contaminants across the air-water interface. Other recent studies have shown that this is the *dominant* atmospheric deposition process for semi-volatile organic contaminants, including PCBs (Baker and Eisenreich 1990; Achman et al. 1993) and low molecular weight PAHs (Nelson et al. 1995). In those studies, the net direction of exchange is often from the water to the air and diffusive gas exchange is large enough to offset wet deposition and dry aerosol deposition.

[13] Because aerosol particle-associated PCBs were present below analytical detection limits, estimates of PCB dry deposition were made using an aerosol PCB level calculated from the measured gaseous PCB concentration and the Junge-Pankow sorption model (Pankow 1987; Leister and Baker 1994).

[14] This is due, in part, to the choice of deposition velocities used for the two studies (0.2 cm/sec for all species in the Great Lakes, 0.26 cm/sec for trace elements and 0.49 cm/sec for organics in the Chesapeake Bay). All of these values are within the generally accepted range for dry aerosol deposition velocities.

FIGURE IV-19
Comparison of Chesapeake Bay and Great Lakes Atmospheric Depositional Fluxes



Source: Baker et al. 1996 (Chesapeake Bay) and Eisenreich and Strachan 1992 (Great Lakes).

CHESAPEAKE BAY TOXIC CONTAMINANT ATMOSPHERIC LOADINGS

To estimate the annual baywide loadings of trace elements and organic contaminants to the Bay, the annual site-specific wet and dry aerosol deposition rates were averaged and the two average rates were multiplied by the surface area of the Bay. Uncertainties in these loading estimates are likely, on the order of a factor of two, mainly due to the inability to estimate dry aerosol loadings on a finer temporal resolution.

Baywide atmospheric loadings of aluminum and iron are estimated at 1,340,000 and 799,000 kg/year, respectively (see Table IV-9). Loadings of trace elements range from 1,110 kg/year for cadmium to 49,400 kg/year for nickel; lead loadings are estimated at 12,500 kg/year. Loading estimates are generally similar for 1991 and 1992, except for nickel and zinc due to

TABLE IV-9
Annual Atmospheric Loadings of Trace Metals and Organic Contaminants to the Chesapeake Bay[a]

Pollutant	Wet Deposition (kg/year)	Dry Deposition (kg/year)	Total Deposition (kg/year)
Aluminum[b]	137,000	1,200,000	1,340,000
Arsenic	607	1,050	1,660
Cadmium	867	240	1,110
Chromium	1,026	2,030	3,060
Copper	5,575	3,620	9,200
Iron	132,800	666,000	799,000
Manganese	13,200	13,600	26,800
Nickel	7,185	6,160	13,300
Lead	5,440	7,080	12,500
Selenium	1,390	2,930	4,320
Zinc	26,000	22,800	49,400
Total PCBs	13	20	37
PAHs			
Anthracene	6	6	13
Benz(*a*)anthracene	9	34	44
Benzo[*a*]pyrene	17	36	53
Benzo[*b*]fluoranthene	36	98	134
Benzo[*e*]pyrene	21	67	88
Benzo[*ghi*]perylene	19	75	94
Benzo[*k*]fluoranthene	22	65	88
Chrysene	29	85	114
Dibenz[*ah*]anthracene	7	16	22
Fluoranthene	70	119	189
Fluorene	16	12	27
Indeno[123*cd*]pyrene	20	78	98
Phenanthrene	63	92	155
Pyrene	75	109	184

[a] To estimate annual baywide loadings of elements and organic contaminants to the entire Chesapeake Bay, the annual site-specific wet and dry aerosol fluxes were averaged and these two average fluxes were multiplied by the surface area of the Bay.
[b] Contribution of aluminum is considered to be entirely from natural sources (i.e., not emitted through human activities).

Source: Baker et al. 1997.

elevated wet deposition measured at the Haven Beach site in 1992. Loadings of PAHs range from 13 kg/year for anthracene to 189 kg/year for fluoranthene. Total PCB loadings are estimated to be 37 kg/year. Interestingly, for many of the pollutants in Table IV-10, wet deposition and dry aerosol depositional fluxes appear to decrease between 1991 and 1992. Whether this reflects a real inter-annual variation or simply results from aggregating measurements from different locations is unclear.

To place these loadings in perspective, they are compared in Table IV-10 to recent estimates of trace metal and organic contaminant loadings delivered to the Chesapeake Bay by the Susquehanna River (Conko 1995; Foster 1995; Godfrey et al. 1995). The Susquehanna River is the largest tributary of the Chesapeake Bay, supplying approximately 60 percent of the freshwater inflow to the estuary. Annual riverine loadings of dissolved and particulate trace metals and organic contaminants were determined by analyzing flow-weighted samples collected at Conowingo, Maryland, between February 1994 and January 1995 (Conko 1995; Foster 1995). Atmospheric deposition directly to the surface waters of the Chesapeake Bay supplies PAH loads that are comparable to or greater than the loads of dissolved PAHs delivered by the Susquehanna River (Table IV-10). Particulate-bound organic contaminants discharged from the river dominate the loading of PAHs, with a large contribution from the high sediment burden carried by the river during high flows. Dissolved total PCB loads from the river are approximately three times those from the atmosphere. Atmospheric depositional fluxes of several elements, including lead, cadmium, and chromium, are within a factor of two of the dissolved load from the Susquehanna River. Again, particulate metal loads from the river dominate over both dissolved riverine loads and atmospheric deposition.

While it is interesting to compare the relative importance of riverine and atmospheric sources of trace elements and organic contaminants to the Chesapeake Bay, the results should be carefully interpreted. While the Susquehanna River delivers large quantities of these substances to the Bay, much of this load is removed in the northern extreme of the Bay (Helz and Huggett 1987) and is delivered episodically during high river flows. Whether particulate-bound metals and organic contaminants are broken down in forms that can be taken up by aquatic organisms is quite unclear. In contrast, atmospheric deposition directly to the water's surface supplies these toxics directly to the water column, without any comparable zone of efficient removal. However, it has recently been suggested that combustion-derived PAHs associated with aerosol particles washed into the surface waters by precipitation also may not be broken down (McGroddy and Farrington 1995). Finally, the distinction between riverine and atmospheric loadings is not clear. Some fraction of the pollutant input from the tributaries results from deposition of atmospheric pollutants to the watershed, with subsequent transmission through the vegetation and soil layers into surface waters (i.e., indirect loading); however, this input cannot yet be quantified.

AREAS OF UNCERTAINTY AND WORK UNDERWAY

Building on the existing CAA requirements, the Chesapeake Bay Program's state and federal partners will focus their efforts on implementation of the Chesapeake Bay Basinwide Toxics Reduction and Prevention Strategy commitment to "establish a more complete baseline and source identification for atmospheric deposition...and set a reduction target from that baseline to be achieved over the next decade" (Chesapeake Executive Council 1994). However, there are several remaining areas of uncertainty to be addressed related to atmospheric deposition of toxic contaminants to Chesapeake Bay. Two significant ones are:

TABLE IV-10
Relative Importance of Sources of Trace Metals and Organic Contaminants to Chesapeake Bay

Pollutant	Susquehanna River Load (kg/year)[a]		Atmospheric Deposition Load (kg/year)[b]
	Dissolved	Particulate	
Aluminum	2,560,000	64,800,000	1,340,000
Arsenic	12,600	ND	1,660
Cadmium	2,130	26,700	1,110
Chromium	4,130	111,000	3,060
Copper	47,800	151,000	9,200
Iron	4,100,000	40,000,000	799,000
Manganese	3,290,000	1,530,000	26,800
Nickel	121,000	65,200	13,300
Lead	6,530	38,600	12,500
Zinc	77,900	360,000	49,400
Total PCBs	97	68	37
PAHs			
Benz[*a*]anthracene	12	364	44
Benzo[*a*]pyrene	5	436	53
Chrysene	15	316	114
Fluoranthene	108	1,020	189
Fluorene	37	85	27
Phenanthrene	63	388	155
Pyrene	104	925	184

[a] Annual loads entering the Chesapeake Bay via the Susquehanna River, measured at Conowingo, Maryland, between February 1994 and January 1995 by Foster (1995) for organic compounds and Conko (1995) for metals.
[b] Total atmospheric deposition loads directly to the surface of the Chesapeake Bay as measured by CBADS.

Source: Baker et al. 1997.

♦ *Dry Deposition*. Dry deposition is viewed as an important mechanism by which chemical contaminants are deposited onto the Bay's tidal surface waters and surrounding watershed. As is the case with nitrogen, there are no widely accepted techniques for direct measurement of dry deposition fluxes of metal or organic contaminants. Although no direct measurements of dry deposition directly to the Bay exist, depositional fluxes have been estimated based on a particle-size-dependent deposition velocity function applied to direct measurements of aerosol concentrations of metals and organic chemical contaminants. Given the absence of direct measures of dry deposition fluxes, there is much uncertainty associated with these loading estimates.

♦ *Transport through the Watershed*. Atmospheric deposition of a pollutant can be a direct input to the Bay surface waters or can be transported from the watershed by surface water and groundwater to the Bay. Transported loads are a component of the total fluvial (i.e., surface water) input from the watershed to the Bay. The degree of landscape retention for a given substance is related to the geomorphology, land use, basic hydrological characteristics unique for each watershed, and soil chemistry. Limited studies to date suggest that the degree of watershed throughput is relatively small (< 30 percent of the rate of rainfall volume). However, evidence to date suggests that watersheds serve as a "reservoir" for atmospherically deposited metals; organically bound metals are sequestered but may be episodically mobilized by acidic precipitation. Because of the relatively large watershed to open water surface area ratio typical of coastal plain estuaries such as Chesapeake Bay (15:1), recent estimates for nitrogen and trace elements suggest that the indirect atmospheric loading may be as significant as the direct input. Thus, while it has been possible to quantify direct atmospheric flux with a fair degree of confidence, one of the primary uncertainties associated with resolving the total atmospheric loading to Chesapeake Bay is in gauging the indirect loading as it relates to the watershed transmission/retention for the myriad of sub-basins (Valigura et al. 1995).

To further improve existing estimates of the relative atmospheric deposition contribution to total chemical contaminant loadings to Chesapeake Bay, the following work is underway. In 1993, the University of Delaware, in cooperation with the U.S. Geological Survey, initiated a pilot study to investigate the transport of atmospherically deposited trace elements through a pristine, forested watershed in the headwaters of the Potomac River (Bear Branch). This study, funded by the Maryland Department of Natural Resources' Power Plant Research Program, has the following specific objectives: (1) to accurately determine the wet and dry atmospheric trace element loads into the watershed, (2) to compare the total atmospheric load versus fluvial output of trace metals and (3) to estimate the transport of atmospherically deposited trace elements through the watershed relative to the trace metals naturally relapsed during weathering of the soil and rock within the study area. The Bear Branch watershed was chosen as it has been well-characterized hydrologically, is representative of the land use in the Potomac basin (60 percent of which is forested), and possesses an unreactive quartzite lithology which simplifies its geochemical weathering behavior. Further watershed transmission studies began in the spring of 1996 in the Appalachian Plateau of Western Maryland. While the results of these studies will represent an initial attempt to quantify the watershed retention/transport of atmospheric loads, further work is needed to extend the study to other regions with differing land use/geomorphology, in order to accurately determine an integrated, baywide watershed transport factor.

The next section describes programs in other coastal waters, as well as research relevant to atmospheric deposition in these coastal waters.

IV.D Coastal Waters

As stated previously, section 112(m) designates "coastal waters" as EPA's National Estuary Program (NEP) and NOAA's National Estuarine Research Reserve System (NERRS) estuaries. These two programs and EPA's Gulf of Mexico Program are the three significant coastal waters programs, outside the Chesapeake Bay Program, established in the last decade. Although the Gulf is not designated by name under section 112(m) of the CAA, 11 estuaries in various locations spanning the Gulf coastline are either NEP or NERRS (current or proposed) designated sites and, thus, are designated Great Waters.

The NEP, NERRS, and Gulf of Mexico programs differ in purpose and procedure, but they all serve to protect and restore the nation's valuable coastal water resources. The remainder of this section provides background information on each of these programs, followed by a discussion of studies of atmospheric deposition to coastal waters and future research needs.

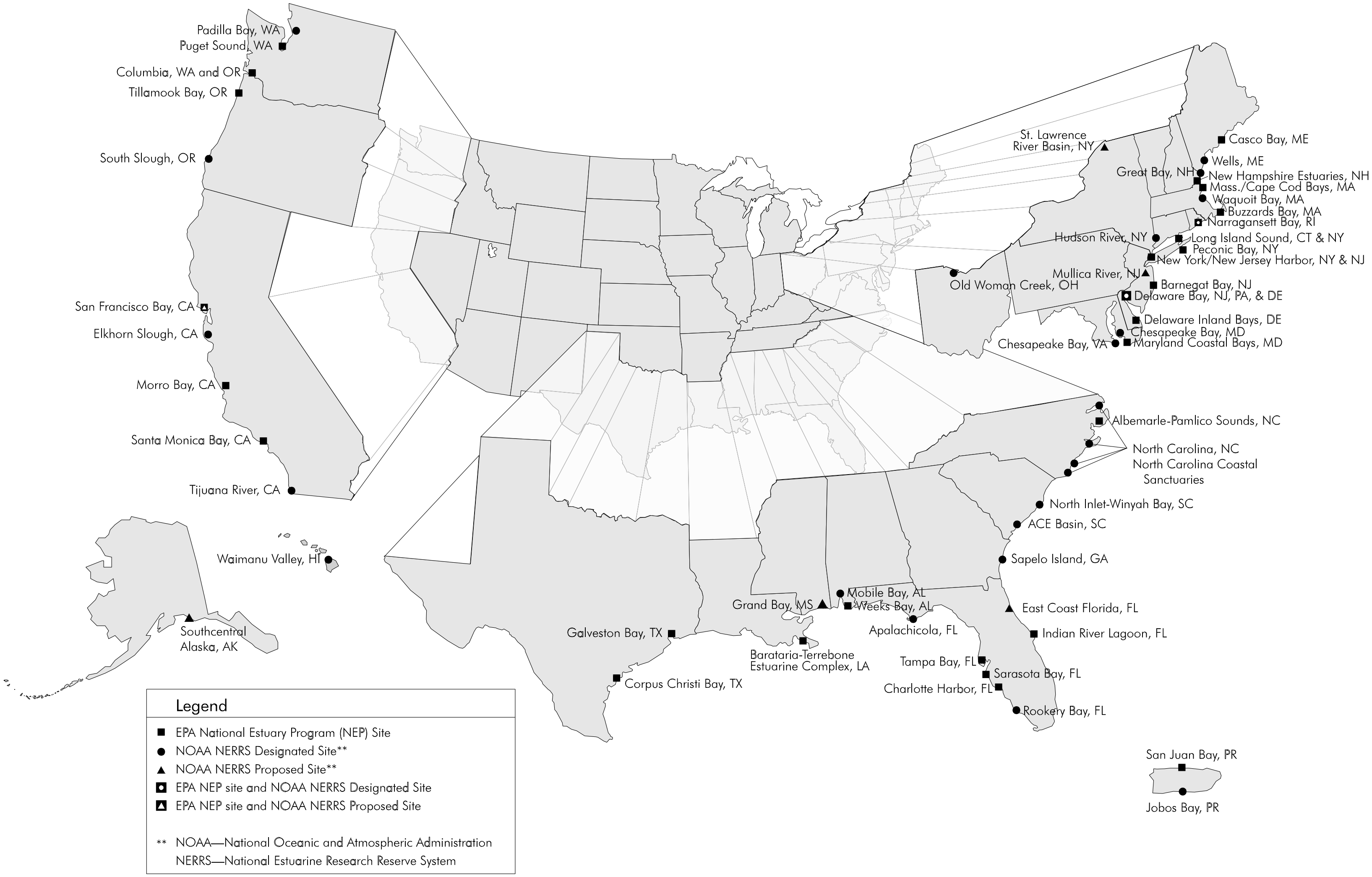
National Estuary Program

Congress established the National Estuary Program (NEP) in 1987 under section 320 of the Clean Water Act. Through the NEP, states nominate estuaries of national significance that are threatened or impaired by pollution, development, or overuse. EPA evaluates the nominations and selects those estuaries for which there is evidence of political support, citizen and government involvement (local, state, regional, federal), and available scientific and technical information to address the identified problems. For the selected estuaries, EPA convenes management conferences with representatives from all concerned groups (e.g., industry, agriculture, environmental organizations, state agencies) to more fully characterize problems and seek solutions through a collaborative decision-making process. Through these conferences, Comprehensive Conservation and Management Plans (CCMPs) are developed, for which EPA provides up to 75 percent of the funding. Each management conference must complete development of the CCMP within three to five years of the date the conference was convened. Upon approval of the CCMP, action plans are carried out by implementation agencies involved with development of the plan.

The purpose of the NEP is to identify nationally significant estuaries, protect and improve their water quality, and enhance their living resources. The NEP currently includes 28 estuaries (individually called NEPs) representing a wide spectrum of environmental conditions (see Figure IV-20). Because there are over 150 estuaries in the United States and only a small fraction can be targeted for action through the NEP, the NEP is intended to act as a national demonstration program, such that results and lessons learned in the NEP estuaries are shared and applied by parties concerned with other estuaries throughout the country. It should be emphasized that the NEP is a management program rather than a research program and relies on the research of other agencies and institutions to support its work. The development of support networks and cooperation between local, state, regional, and federal agencies is one of the program's greatest assets.

Several NEPs have identified atmospheric deposition of pollutants as a concern to the health of their estuaries. These NEPs have either initiated studies on the contribution of atmospheric deposition to annual loadings for nitrogen and/or other pollutants or expressed serious interest to EPA in conducting such projects. Nine NEPs submitted proposals to EPA in early 1996 for funding under section 112(m) of the CAA. To date, only two NEPs (Tampa

FIGURE IV-20
Locations of NEP and NERRS Sites



Bay and Galveston Bay) have received funding under section 112(m) to conduct such work. Other NEPs (Casco Bay, Delaware Bay, Long Island Sound, Massachusetts Bays, and Peconic Bay) have initiated exploratory studies funded by their own program and other sources. Atmospheric deposition research related to the NEPs is discussed later in this section.

National Estuarine Research Reserve System

Another program established to recognize the importance of estuaries is the National Estuarine Research Reserve System (NERRS), which was created by Congress in 1972 under the Coastal Zone Management Act and which operates under the authority of NOAA. The mission of NERRS is to establish and manage, through the cooperation of federal, state, and community efforts, a national system of estuarine research reserves that are representative of various regions and estuary types in the United States, in order to provide opportunities for long-term research, education, and stewardship.

The process for designating and maintaining a NERRS site includes five main activities, all of which may be partially funded by NOAA: (1) predesignation phase (includes selection of the site by the state and, after approval of the site by NOAA, preparation of a draft and final management plan and environmental impact study and completion of basic characterization studies); (2) acquisition of land and development activities; (3) after designation as an NERRS site, implementation of research, educational, and research programs detailed in the research reserve management plan; (4) estuarine research and monitoring; and (5) educational and interpretive activities.

Currently, 22 areas are designated as NERRS sites, including portions of Chesapeake Bay and associated lands in Maryland and Virginia (see Section IV.C for a detailed discussion of Chesapeake Bay). Six additional NERRS sites have been proposed or are in the beginning stages of development. See Figure IV-20 for the location of the NERRS estuaries.

Studies on the direct contribution of atmospheric deposition to NERRS waters are limited at this time. Available information on atmospheric deposition research related to NERRS waters is presented later in this section. For example, the indirect contribution of atmospherically deposited nitrogen to Waquoit Bay, Massachusetts, through its watershed has been estimated and modeled as part of the multi-year Waquoit Bay Ecological Risk Assessment Case Study (NOAA and MA DEM 1996; U.S. EPA 1996f).

Gulf of Mexico Program

The Gulf of Mexico is a very important resource to all of North America. Its surface area is about 1,603,000 km^2, large enough to cover one-fifth of the continental United States. The U.S. portion of the Gulf's shoreline measures over 2,500 km from the Florida Keys to the Rio Grande. Taking into account the shoreline length of all the bays, estuaries and other coastal features of the Gulf, its effective shoreline length is about ten times that amount. The 21 major estuaries along the Gulf coast account for 24 percent of all estuarine area in the 48 contiguous states, and 55 percent of the marshes. The watershed of the Gulf includes more than two-thirds of the continental United States (plus one-half of Mexico and parts of Canada, Guatemala, and Cuba), with the Mississippi River watershed alone draining about 40 percent of the continental United States.

The Gulf of Mexico Program (GMP) was established in 1988 in response to citizens' concerns over declines in the Gulf's fish, shellfish, and wildlife; the quality of life in many coastal communities; the need to protect the remaining valuable resources and prevent problems before they occur; and to forge a positive relationship between ecological health and economic vitality of the Gulf region. The GMP is a unique organization that involves representatives from government agencies (federal, state, and local), business and industry, non-profit organizations and educational institutions, and interested individuals in the process of setting environmental goals and implementing actions to achieve those goals. The aim is to foster coordination and cooperation among these organizations in order to reduce costs and coordinate actions.

The GMP is not a regulatory program, but rather an approach to environmental protection, similar to the Chesapeake Bay Program, that is founded on the principles of:

- Partnership among government agencies, private, and non-government interests to define and characterize concerns and implement solutions;

- Sound science and information as the basis of informed decision-making to guide actions; and

- Public involvement to determine goals, identify solutions, and generate the consensus needed for action.

Since its beginning, the GMP has made significant progress in effectively involving a broad spectrum of the public in defining goals and objectives and in characterizing fundamental issues. The fundamental goals of the GMP are to:

- Protect human health and the food supply;

- Maintain and improve Gulf habitats that support living resources (fish, shellfish, and wildlife); and

- Maintain and enhance the sustainability of the Gulf's living resources.

In the past few decades, the Gulf of Mexico has been degraded, largely due to nutrient enrichment and habitat loss. The contribution of atmospheric nitrogen to nutrient enrichment is not well understood and is possibly a significant concern. Fed by nutrient-enriched waters of the Mississippi River, a large area of near-bottom waters commonly become depleted in oxygen, or hypoxic. At its peak, this area (known as the "hypoxic zone") can extend over a 18,192 km^2 area from the coastal waters of the Mississippi River Delta of Louisiana to those of eastern Texas. Stresses to the benthic (bottom-dwelling) community have been observed in this zone, including mortality of larger non-swimming benthic organisms. This and other possible disruptions to the food chain threaten to affect the commercial and recreational fish species within the hypoxic area. In addition to the Louisiana Shelf hypoxic zone, 18 other coastal areas in the Gulf have experienced hypoxia due to increasing nutrient concentrations or loads. Evaluation of atmospheric deposition of pollutants to the Gulf is discussed below, including research in NEPs located in the Gulf, as well as two studies conducted in the Gulf as a whole.

Studies of Atmospheric Deposition in NEP and Other Coastal Waters

Nitrogen Loadings to Coastal Waters

At least 40 studies around the world, the majority of which have been published since 1990, have addressed at least the direct loading component of atmospheric nitrogen loadings. However, the measurement and modeling techniques used vary considerably among individual studies, making comparisons difficult. Table IV-11 presents a summary of selected studies performed along the East and Gulf coasts of the United States that are comparable in broad terms. The two criteria for selecting these studies were that the study results were either published in a credible peer-reviewed journal or advocated by a major management organization (e.g., an EPA NEP). These studies can be divided into two groups: those that considered both direct and indirect nitrogen loads and those that considered only direct loads. Data from these studies show that, in general, the amount of atmospheric nitrogen input is related to the size of the waterbody and its watershed. To some extent, the percent load from atmospheric deposition is influenced by whether both direct and indirect deposition were considered.

Municipal and industrial wastewater discharges and urban runoff/storm sewer inputs have historically been considered the largest sources of pollutants to coastal waters. Recently, however, researchers have begun to investigate the role of atmospheric deposition as a source of pollutants in a coastal waters (e.g., Paerl 1985, 1993; Scudlark and Church 1996). Assessing the impact of atmospheric deposition of pollutants has become a priority for many NEPs and other coastal watershed protection programs. There is a clear need to characterize the types, quantities, and sources of pollutants that are being directly and indirectly deposited from the atmosphere into these estuaries. Recent studies on atmospheric deposition to coastal waters are discussed briefly below and are presented in Tables IV-12 and IV-13. Data as of December 1995 on the contribution of atmospheric deposition to nitrogen loadings to Chesapeake Bay and other coastal waters are presented in Table IV-11; in this table, information is presented first for Chesapeake Bay and a few related tributaries, followed by other coastal waters in descending order of tidal water area. Data on the contribution of atmospheric deposition to the loadings of toxic pollutants in coastal waters are presented in Table IV-12; the coastal waters are listed in geographical order clockwise from the northeast coast to the northwest coast. In general, the studies discussed below have evaluated the relative contribution of nitrogen and other pollutants of concern, and do not attempt to identify the particular emission sources contributing to this pollution.

As mentioned above, research on atmospheric deposition to Tampa Bay, Florida, and Galveston Bay, Texas, has been conducted under the Great Waters program. The Tampa Bay and Galveston Bay studies are discussed first, followed by a description of other studies of atmospheric deposition to NEP estuaries (i.e., those that have been funded through sources other than EPA's Great Waters program). Initial observations from these studies suggest that direct and indirect loadings from air deposition may be significant sources of nitrogen and toxic pollutants to coastal waters.

Tampa Bay. As recently as 1991, atmospheric deposition of nitrogen, air toxics, and other pollutants was assumed to have a minimal effect on water quality in Tampa Bay. However, based on a methodology developed by the Environmental Defense Fund (Fisher et al. 1988), early calculations provided an early indication of likely nitrogen loadings from atmospheric deposition in Tampa and Galveston Bays.

TABLE IV-11
Estimates of Atmospheric Nitrogen Loadings to Selected Coastal Waters[a]
(in millions of kg)

Coastal Water	Surface Area (km^2)		Deposition to Watershed	Direct Deposition to Tidal Waters	Indirect Atmos. Load From Watershed	Total Atmospheric Load	Total Load From All Sources	% Load from Atmosphere	Reference[b]
	Watershed	Tidal Waters							
Chesapeake Bay (MD/VA)	165,886	11,400	175	16	29	45	170	27	5
Rhode River (MD)	33	4.9	—	0.005	—	0.005	0.012	40	6
Choptank River (MD)	1,779	361	—	0.57	—	0.57	1.54	37	11
Patuxent River (MD)	2,393	137	—	0.22	—	0.22	12.6	13	11
Potomac River (MD)	29,940	1,210	—	1.9	—	1.9	35.5	5	11
New York Bight (NY/NJ)[c]	50,107	38,900	69	54	8	62	164	38	1
Albemarle-Pamlico Sound (NC)	59,197	7,754	~39	3.3	6.7	10	23	44	4
Long Island Sound (NY/CT)	43,481	4,820	43	5	6	11	60	20	3
Massachusetts Bays (MA)	—	3,700	—	1.6-6	—	1.6-6	22-30	5-27	15
Delaware Bay (DE)	36,905	1,846	53	3	5	8	54	15	2
Tampa Bay (FL)	6,216	1,031	—	1.1	—	1.1	3.8	28	14
Guadalupe Estuary (TX)	—	551	—	0.31	—	0.31	4.2-15.9	2-8	13
Narragansett Bay (RI)	4,708	328	4.2	0.3	0.3	0.6	5	12	1
			—	0.4	—	0.4	9	4	12
Newport River Coastal Waters (NC)	340	225-1,600	—	0.095-0.68	—	0.095-0.68	0.27-0.85	36-80	4
Sarasota Bay (FL)	524	135	—	0.16	—	0.16	0.6	26	10
Delaware Inland Bays (DE)	800	83	—	0.28	—	0.28	1.3	21	9
Flanders Bay (NY)	83	39	—	0.027	—	0.027	0.36	7	8
Waquoit Bay (MA)	~70	~8	0.062	—	0.0065	0.0065	0.022	29	7

[a] Estimates as of December 1995.
[b] (1) Hinga et al. 1991; (2) Scudlark and Church 1993; (3) Long Island Sound Study; (4) Paerl and Fogel 1994; (5) Linker et al. 1993; (6) Correll and Ford 1982; (7) Valiela et al. 1996; (8) Peconic Bay NEP; (9) Delaware Bays NEP; (10) Sarasota Bay NEP 1995; (11) Boynton et al. 1995; (12) Nixon et al. 1995; (13) Brock et al 1995; (14) Tampa Bay NEP, Zarbock et al. 1994; (15) Massachusetts Bays NEP 1996.
[c] New York Bight extends from Cape May, New Jersey, to Long Island Sound.

Source: Adapted from Valigura et al. 1996.

TABLE IV-12
Studies of Atmospheric Loadings of Toxic Pollutants to NEP Coastal Waters

Coastal Water	Pollutants of Concern Evaluated[a]	Relative Contribution of Atmospheric Deposition for the Great Waters Pollutants of Concern	Reference
Massachusetts Bays (MA)	PAHs, PCBs, cadmium, lead, mercury	Direct atmospheric deposition estimated to contribute: PAHs, 9-46%; PCBs, 28-82%; cadmium, 17-31%; lead, 39-45%; mercury, 4-13%.	Menzie-Cura & Associates 1991
Narragansett Bay (RI)[b]	PCBs, PAHs	Direct atmospheric deposition found to contribute 3% of PCBs and 12% of PAHs.	Latimer 1997
New York-New Jersey Harbor Estuary and Bight (NY/NJ)	Cadmium, lead, mercury, PCBs, dioxins, PAHs, various pesticides	Atmospheric deposition identified as a significant contributor to total pollutant loading for lead (39-54%), but may have been over-estimated. For other pollutants, either atmospheric deposition was insignificant or estimates were not developed.	NY-NJ Harbor NEP 1995
Delaware Bay (DE/NJ/PA)[b]	Lead, mercury, PCBs, various pesticides, volatile organic compounds (VOCs)	Atmospheric deposition (both direct and indirect) found to be a significant source of mercury (80%) and PCBs (34%). For lead, atmospheric deposition contributed less than 5%. For other pollutants, either atmospheric deposition was insignificant or estimates were not developed.	Frithsen et al. 1995b
Tampa Bay (FL)	Cadmium, lead, mercury, chlordane, DDT, dieldrin, PCBs, PAHs	Direct and indirect atmospheric deposition identified as a significant contributor of cadmium (46%), lead (12%), and PCBs[c], but not a significant source of chlordane, DDT, dieldrin, or mercury (1% each). Estimates for PAHs were not developed.	Frithsen et al. 1995a
	Cadmium, lead, mercury, chlordane, DDT, dieldrin, PCBs, PAHs	On-going study - no results yet.	U.S. EPA 1995f
Galveston Bay (TX)	PAHs, PCBs, selected pesticides, lead, cadmium, mercury	On-going study - no results yet.	U.S. EPA 1995g
Santa Monica Bay (CA)	PAHs, metals, chlorinated organics	Atmospheric deposition was estimated to be a significant source of lead and PAHs.	SMBRP 1994

[a] For a discussion of other pollutants evaluated, study methods, and uncertainties, see referenced study.
[b] These NEPs also are NERRS designated sites.
[c] Estimates of PCB loadings could be made for atmospheric deposition only; therefore, a relative comparison to other sources could not be made.

A recent study of nutrient (i.e., nitrogen and phosphorus) and suspended solids loadings conducted for the Tampa Bay NEP suggests that direct atmospheric deposition of nitrogen to the tidal waters of Tampa Bay is the second largest source of nitrogen entering the Bay, contributing up to 28 percent of the total nitrogen load (Zarbock et al. 1994). The largest source of nitrogen, according to that study, is urban storm water runoff. A portion of the nitrogen entering the Bay from urban storm runoff represents atmospherically deposited nitrogen to impervious surfaces such as paved roads and sidewalks. These studies prompted the Tampa Bay NEP to revise its CCMP to consider atmospheric deposition issues.

Another study, conducted by the Tampa Bay NEP (Frithsen et al. 1995a), investigated the contribution of atmospheric deposition relative to point sources, urban runoff, and other nonpoint sources for specific chemical contaminants of concern. The contaminants, which were selected based on their potential for toxic effects and their concentrations observed in sediment samples, included: six metals (cadmium, chromium, copper, lead, mercury, and zinc); four organochlorine pesticides (chlordane, DDT, dieldrin, and endrin); and two classes of organic chemicals (polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs)). In addition, loadings were estimated for arsenic and iron because of the potential of these chemicals have for environmental harm or interaction with contaminants of concern through chemical processes. Because the study used numerous information sources, representing a wide range of spatial and temporal conditions, a great deal of uncertainty exists regarding the absolute estimates of atmospherically deposited chemical contaminants to Tampa Bay. Study results are intended to establish the relative magnitudes of different classes of sources in order to set priorities for more detailed research and monitoring activities. Some general conclusions, however, are apparent:

- Contaminant inputs to Tampa Bay from runoff and transfer of atmospherically deposited contaminants to the watershed (i.e., indirect loading) are approximately two-thirds the contribution of contaminant inputs from direct deposition to tidal waters (i.e., direct loading);

- Atmospheric deposition contributes a sizeable percentage to total annual load for the following metals: cadmium (46%), copper (18%), chromium (13%), lead (12%), and iron (11%);

- Atmospheric deposition of mercury is around 4 kg/yr and atmospheric deposition of pesticides is estimated as 10 kg/yr, each representing about one percent of the total load; and

- Atmospheric deposition is the only pathway that contributed a measurable amount of PCBs, estimated as a minimum total load of 11 kg/yr. No estimate could be developed for PAHs using available datasets.

Ongoing monitoring, described below, conducted by the Tampa Bay NEP, local governments, and other collaborators will better define the spatial distribution of atmospherically deposited chemical contaminants and nitrogen throughout the 10 major basins of Tampa Bay and its watershed as well as the relative contributions of local, regional, and global emission sources.

An ongoing cooperative study administered by the Tampa Bay NEP will substantially expand air transport and deposition monitoring and modeling projects in an effort to develop nationally recognized quantitative assessments for important air deposition parameters. Now in its second year of operation, the Tampa Bay Atmospheric Deposition Study (TBADS) involves EPA's Great Waters program; the local governments in Hillsborough, Pinellas, and Manatee Counties; the Florida Department of Transportation; the Southwest Florida Water Management District; Florida State University; a private consultant; and other organizations. TBADS will attempt to determine: (1) what fraction of the nitrogen and toxic pollutants emitted annually by specific sources within the Tampa Bay watershed enter the Bay waters (i.e., are deposited either onto the water surface directly or onto the watershed and subsequently enter the Bay waters through runoff); and (2) what are the relative contributions of local sources (i.e, inside the watershed) versus remote sources (i.e, outside the watershed) to atmospherically deposited nitrogen and toxic pollutants in the watershed. Projects that have been initiated to address elements of this air deposition program include:

- Intensive daily wet and dry deposition monitoring at Gandy Site, located on Tampa Bay's Interpeninsula, for nitrogen and toxic pollutants for an additional 12 months, yielding two years of continuous data collected and analyzed according to AIRMoN protocol;

- Application (with the Florida State University Center for Tropical Meteorology) of a regional air mass movement model developed by Pennsylvania State University (Penn State/NCAR Mesoscale Model Version 5) to investigate air transport at a coarse grid for the southeastern United States while maintaining a much higher resolution grid over the Tampa Bay area to estimate retention times for air masses within the watershed under different meteorological conditions;

- Integration of NOAA's Physical Oceanographic Real-Time System (PORTS) Meteorological data collected at several over-water stations (with NOAA and the University of Florida); and

- Intensive stormwater sampling to measure stormwater runoff, nitrogen, and toxic contaminant concentrations and loads at up to four gaged subbasins. Data will be used to estimate the relative contribution of atmospheric loading to stormwater for different land use types (residential, urban, industrial, and/or urban parks); it is expected that this transfer coefficient information will be useful not only to Tampa Bay and Florida, but also to watersheds nationwide.

An important element of the Tampa Bay atmospheric deposition program is the participation and coordination of local and federal government programs and state agencies with the Great Waters program and the Tampa Bay NEP.

Galveston Bay. The Great Waters program conducted a screening atmospheric deposition monitoring program in Galveston Bay, Texas, which was chosen as the site to establish monitoring for the Texas Regional Integrated Atmospheric Deposition Study (TRIADS) as a representative of a Gulf of Mexico estuary. Monitoring at the TRIADS site began in February 1995. To facilitate comparability, the sampling and analytical design of TRIADS is similar to that of existing monitoring sites in the Great Lakes and Chesapeake Bay. The goals of this study are to evaluate the contribution of atmospheric deposition of selected contaminants to the Bay and to evaluate long-range transboundary transport of contaminants. Pollutants measured include cadmium, lead, mercury, nitrogen, PAHs, PCBs, and selected pesticides. Results from TRIADS complement and add to data from other investigations in Galveston Bay, including studies by the Galveston Bay NEP, EPA's Environmental Monitoring and Assessment (EMAP) program, NOAA National Status and Trends (NS&T) program, and special urban-pollutant studies in Houston, Texas. Data from these programs and the TRIADS data will be used to estimate the cumulative, direct and indirect impacts of atmospheric deposition to pollution of Galveston Bay.

Although early calculations suggested that atmospheric deposition could be a significant contributor to nitrogen loads delivered annually to Galveston Bay, the relevance of this finding to the health of the ecosystem for Galveston Bay is not as obvious as for either Chesapeake Bay or Tampa Bay. While all three estuaries have experienced declines in submerged aquatic vegetation (SAV), studies on SAV in Galveston Bay are limited in contrast to documented cases of large-scale changes in other major estuaries (Pulich et al. 1991). While atmospheric nitrogen loading may contribute to the incidence of hypoxia, other factors appear to be causing this phenomenon, which is quite localized in Galveston Bay compared to its manifestation in the other two estuaries.

In contrast to the perceived limited biological effects from atmospheric deposition of nitrogen, previous research has suggested that atmospheric deposition of toxic contaminants may be affecting fish and shellfish in Galveston Bay, and thus contributing to human health risk. A pilot study performed for the Galveston Bay NEP documented the presence of dioxins, furans, lead, mercury, PAHs, PCBs, and pesticides in certain species of finfish and shellfish, but could not determine the sources of these contaminants (Brooks et al. 1992). Monitoring data from the TRIADS site detected the presence of all these chemical contaminants in air samples, suggesting that atmospheric deposition may be a significant source (Battelle 1995). Continued monitoring will enable scientists and managers to more fully evaluate this problem and determine the relative effect of atmospheric deposition versus point and nonpoint source inputs into Galveston Bay.

Casco Bay. The primary pollutants of concern for atmospheric deposition to Casco Bay, Maine, include PAHs, PCBs, nitrogen, phosphorus, sulfates, pesticides, and mercury and other trace metals. Recent sediment studies have found elevated concentrations of some pollutants (i.e., cadmium, lead, mercury, PAHs, PCBs, silver, and zinc) near population centers and waste discharges, but also observed elevated levels in rural eastern Casco Bay away from these known sources (Wade et al. 1995). A circulation model study of the Bay did not clearly indicate any possible sources for these pollutants, suggesting atmospheric deposition as a significant source (Pearce et al. 1994). While elevated levels of lead found in Casco Bay sediments were relatively near potential sources, elevated levels of cadmium were found far from any known local source. A deposition study would provide empirical verification of processes believed to be occurring at Casco Bay.

Delaware Bay. In Delaware Bay, studies have shown that direct and indirect atmospheric deposition provide 15 percent of the annual nitrogen input, increasing to 25 percent in late spring and early summer (Scudlark and Church 1993). The relative nitrogen loading is slightly lower than observed in nearby Chesapeake Bay (27 percent), and much lower than in the Delaware Inland Bays (Rehoboth and Indian River Bays) where direct atmospheric deposition alone contributes 27 percent of the total nitrogen load (Cerco et al. 1994). The contribution to the Delaware Bay is lower because of higher point source nitrogen loading to Delaware Bay and the influence of a highly urbanized watershed.

As part of a Delaware Estuary Program study to estimate contaminant inputs, atmospheric deposition was found to be a significant source of mercury (80 percent) and PCBs (34 percent) (Frithsen et al. 1995b). As is the case in other regions, more research is warranted on atmospheric inputs of mercury and the resulting effects on estuarine and human health. To evaluate the effect atmospheric deposition of mercury has on the Delaware, Rehoboth, and Indian River estuaries, a precipitation monitoring station was established at Lewes, Delaware, in 1995 in conjunction with EPA's National Atmospheric Deposition Program Mercury Deposition Network.

Hypoxia Conference

A conference was convened in 1996 in response to the hypoxia problem in the Gulf of Mexico. Topics addressed included:

- Characterization of the hypoxic zone;
- Economic impacts and trends in fisheries attributable to the hypoxia;
- Causes of the hypoxic zone;
- Sources and delivery of nutrients in the watershed, including atmospheric deposition;
- Current efforts to control nutrient loads; and
- Information and policy required for action.

Gulf of Mexico. It is probable that significant amounts of nitrogen are deposited into the Mississippi River Basin via atmospheric deposition, but there has been little investigation conducted regarding atmospheric nitrogen as a source of nitrogen for the Mississippi River drainage basin. Some basic estimates using National Atmospheric Deposition Program (NADP) data were provided at a Hypoxia Conference held by the Gulf of Mexico Program in August 1996. These estimates showed significant variability in quantity of atmospherically deposited nitrogen, with a range of 0.55 million to 3.08 million tons. This variability is due to differing assumptions of what atmospheric nitrogen input sources should be included, what forms of deposition should be measured (e.g., dry deposition), and what nitrogen compounds should be analyzed (e.g., ammonium). At the high end, atmospheric nitrogen would be on par with animal manure, ranking as the second highest source of nitrogen input to the watershed. At the lower end, estimated atmospheric nitrogen inputs would rank as the fifth highest source of nitrogen input for the watershed. This wide variability in estimated quantity points to the need for further and more refined estimates of atmospheric nitrogen inputs to the Gulf.

Long Island Sound. A chronic problem in Long Island Sound is the low oxygen levels (hypoxia) that are observed during the summer. An early study noted that excess nitrogen loading was a major cause of hypoxia and estimated that atmospheric loading directly to the water surface contributed 8 percent of the total nitrogen delivered to the Sound (LIS Study 1990). A later study produced essentially the same estimate for the total contribution of direct atmospheric deposition, but divided it into two components: an amount from "natural" sources (i.e., background approximating "amount believed to have been delivered to Long Island Sound in pre-Colonial days") and an amount from "human-induced" sources (LIS Study 1994). Using measurements of wet and dry deposition from two sites along the Connecticut shore, Miller et al. (1993) estimated direct atmospheric loadings to Long Island Sound. The Long Island Sound Study used these data and literature values to develop the estimates shown in Table IV-12, and concluded that (direct and indirect) atmospheric deposition may be responsible for 17 to 24 percent of total nitrogen entering the Sound.

In the most recent study, all sources of nitrogen, including atmospheric deposition, are divided into "natural" and "human-caused" components (Stacey 1996). This study concluded that atmospheric deposition from human activities in the New York and Connecticut portions of the Long Island Sound watershed accounts for 13.6 percent of the total enriched or "human-caused" load. Further work is necessary to model relationships among air quality, direct and indirect atmospheric deposition, and runoff concentrations to receiving waters of the Sound.

Massachusetts Bays. In one Massachusetts Bays NEP study, direct atmospheric deposition was estimated to contribute 5 to 16 percent of total nitrogen load to Massachusetts Bays (Menzie-Cura & Associates 1991). In another Massachusetts Bays NEP study, direct atmospheric deposition of nitrogen was estimated to account for 6 to 8 percent of total nitrogen loadings to the Bays (Zemba 1995). Different methodologies were used to estimate nitrogen loadings in these two studies. The estimate by Zemba (1995) used literature values and ten years (1981-1991) of wet deposition data from the NADP. Other studies cited by the Massachusetts Bays NEP suggest that the contribution of atmospheric deposition may be higher, about 16 to 20 percent of total nitrogen load, excluding exchange with the Gulf of Maine (Massachusetts Bays NEP 1996).

Atmospheric deposition is also a significant contributor of organic pollutants and trace metals to Massachusetts Bays. Menzie-Cura (1991) estimated that direct atmospheric deposition was a significant source of PAHs (9-46 percent), PCBs (28-82 percent), cadmium (17-31 percent), and lead (39-45 percent). A subsequent Massachusetts Bays NEP study generally corroborated the Menzie-Cura (1991) metal deposition results, although lead deposition rates were slightly lower (Golomb et al. 1995). The lead deposition estimates may be lower in Golomb (1995) because the data used in the Menzie-Cura study were obtained prior to the phase out of leaded gasoline. Golomb (1995) also indicated that PAH deposition may have been underestimated and that PCB concentrations were below detection limits and, therefore, atmospheric deposition rates for PCBs were not calculated. Because PCB concentrations were below the detection limit, more precise field measurements of wet and dry deposition of PCBs are necessary to verify the initial estimates and to determine the relative impact of atmospheric deposition of PCBs to Massachusetts Bays.

Peconic Bay. Nitrogen from atmospheric deposition to the Peconic River and Flanders Bay, New York, is estimated to be about 73 kg/day, or about five percent of the total nitrogen loading to that area (Suffolk County 1992). The impact of atmospheric deposition on eutrophication in Peconic River and Flanders Bay is considered to be relatively small in relation to other point and nonpoint sources. Atmospheric deposition is believed to be much more significant in terms of relative eutrophication impacts to Peconic Estuary surface waters east of Flanders Bay. Detailed loading estimates for these eastern areas, as well as for specific subwatersheds, are being developed for the Peconic NEP waters, and the relative eutrophication impacts of sources are being assessed through computer modeling.

Future Research Needs in NEP and Other Coastal Waters

Research on atmospheric deposition to coastal waters has been limited to a few areas, and most studies have focused on identifying and determining the concentration of pollutants of concern in water and sediment, and measuring concentrations of pollutants in precipitation. Due to limited funding, many preliminary NEP studies are restricted to using historical data to estimate atmospheric deposition. Some NEP studies have used a rough mass balance approach to determine the relative loading of each pollutant to the estuary, but more precise quantitative mass balances are needed, which require accurate and comprehensive atmospheric data.

Establishing the total contribution of pollutants and their sources is an important part of developing and implementing CCMPs for NEPs, and the lack of knowledge about the concentrations, deposition, and potential sources of airborne pollutants makes sound policy formation for the estuaries difficult. The question of the magnitude of pollutant deposition from the air has become more important as other sources of pollution to rivers, lakes, streams, and coastal waters have been identified and significantly reduced.

Research questions for the NEP estuaries and other coastal waters include:

- What are the concentrations and loadings of pollutants that are being supplied by atmospheric deposition?

- What are the relative contributions of these inputs to the total load of pollutants entering the estuary?

- What are the emission sources that affect the estuaries and where are they geographically located?

- Does atmospheric deposition (direct and indirect) of contaminants cause or contribute to biological harm in benthic (bottom-dwelling) or pelagic (suspended, planktonic, or water column) communities, or affect human health?

- What economically and technically feasible methods are available to effectively reduce airborne pollutants and their effects on estuaries?

The NEP estuaries provide an excellent opportunity to evaluate the effects and contribution of atmospheric deposition of contaminants to a varied set of ecological, environmental, and anthropogenic conditions. The NEP also provides a "grassroots" forum for addressing and correcting regional and national air quality issues as they pertain to our coastal waters. Recommendations for further atmospheric deposition research in coastal waters to help answer the above questions include:

- Utilize existing databases and ongoing work of established research programs and coordinate research initiatives with these programs;

- Protect and enhance existing monitoring programs;

- Establish long-term water and air quality monitoring programs that incorporate sampling for atmospheric deposition of contaminants for a subset of NEP estuaries representing various geographical regions and environmental conditions;

- Use sampling data from monitoring programs to track trends and spatial variability to develop more accurate loading estimates;

- Coordinate efforts between NEP estuaries and other Great Waters program studies to identify local, regional, and national sources of airborne pollutants;

- Pursue detailed atmospheric chemistry and deposition models for estimating atmospheric deposition to NEP estuaries;

- Develop a multi-party effort to identify and demonstrate appropriate pollution prevention techniques;

- Apply existing atmospheric circulation models to fill in data gaps between measured and estimated atmospheric deposition and to aid in tracing the pollutants in the estuaries back to their probable sources; and

- Support process-related research to establish cause and effect relationships between atmospheric deposition of contaminants and alterations of water quality, fisheries, recreational and other economic and ecological resources of receiving estuarine and coastal waters.

This research is needed not only to assist decision-makers for specific coastal waters, but to form a comprehensive picture of atmospheric deposition across the United States. In addition, coordinated use of other mechanisms, such as voluntary pollution prevention, can help control the negative impacts of atmospheric deposition to water quality in NEP estuaries, especially at the local and regional level.

CHAPTER V
CONCLUSIONS AND FUTURE DIRECTIONS

This report affirms and provides added support for the findings of the First Report to Congress that, for studied Great Waters, atmospheric deposition of toxic pollutants and excessive nitrogen is often an important factor affecting the environmental conditions of these waterbodies and can contribute to adverse ecological and human health effects. Moreover, the contribution of atmospheric deposition can be significant as part of the total loading for many waterbodies. For the freshwaters, such as the Great Lakes and Lake Champlain, metals, organic compounds, and pesticides released into the atmosphere have been measured in significant quantities near these waterbodies, and their deposition has been measured or calculated. For Chesapeake Bay and many other U.S. coastal waters, the impact of atmospheric deposition is not only from toxic pollutant releases, but also from inputs of nitrogen compounds that contribute to eutrophication (an overabundance of nutrients in a waterbody). Significant atmospheric loadings for both nitrogen and toxic pollutants have been determined for certain estuaries.

Since the First Report to Congress, significant progress has been made to increase our knowledge of atmospheric deposition to the Great Waters (see sidebar summary). As highlighted in this report, greater attention has been placed on monitoring and modeling atmospheric deposition at the individual Great Waters, with many of these studies funded or supported by EPA's Great Waters program. Quantitative information has continued to be gathered on the atmospheric levels of pollutants and their deposition to the Great Waters. Monitoring studies have been conducted to provide waterbody-specific data on deposition and the relative contribution of atmospheric deposition to total loadings, as well as to develop and improve measurement and modeling methods. Moreover, the findings from the investigation of one waterbody are expected to lead to a more informed or efficient assessment of atmospheric deposition and its effects for other waterbodies.

Progress Since the First Report to Congress in Cooperation With the Great Waters Program

- Initiation of a comprehensive mass balance model of Lake Michigan, for which results will estimate relative loadings, with the methodology to be adapted and applied to other waterbodies;
- Improved and refined source emission inventories for pollutants of concern;
- Formal policy commitment of the U.S. and Canadian governments to address Virtual Elimination of Persistent Toxic Substances in the Great Lakes;
- Increased monitoring and modeling studies in progress at the specific waterbodies;
- State-of-the-art air quality modeling of nitrogen sources, transport, and deposition to the Chesapeake Bay watershed;
- Increased activities at the coastal waters including development and implementation of monitoring studies, comprehensive pollution prevention plans, and management programs;
- Understanding that atmospheric deposition contributes significantly to excessive nitrogen loads in 13 of the 14 studied estuaries along the East and Gulf coasts; and
- Numerous programs and activities that are funded and supported by the Great Waters program.

Based on the scientific information currently available, EPA continues to support the three broad conclusions presented in the First Report to Congress regarding potential adverse effects, relative atmospheric loadings, and sources of atmospheric deposition.

- ♦ Adverse effects (e.g., cancer and developmental effects) that the Great Waters pollutants of concern can cause in humans and wildlife are fairly well understood. However, data are insufficient at this time to establish the linkage between atmospheric deposition of these pollutants and adverse effects.

- ♦ Atmospheric deposition can be a significant contributor of toxic chemicals and nitrogen compounds to the Great Waters. The relative importance of atmospheric loading for a particular chemical in a given waterbody depends on many factors, including characteristics of the waterbody, properties of the chemical, and the kind and amount of atmospheric or water discharges (airborne or waterborne).

- ♦ Airborne emissions from local as well as distant sources contribute pollutant loadings to waters through atmospheric deposition. Determining the relative roles of particular sources — local, regional, national and possibly global — that contribute significant deposition to specific waterbodies is complex, requiring careful monitoring, atmospheric modeling, and other analytical techniques.

In addition to these conclusions, EPA also <u>reaffirms its support for the three major strategic themes</u> developed in the First Report to Congress, which provide a broad scope for recommendations for action.

(1) EPA will continue ongoing efforts to implement section 112 and other sections of the CAA and use results from this report in the development of policy that will reduce emissions of the Great Waters pollutants of concern.

(2) EPA recognizes the need for an integrated multimedia approach to the problem of atmospheric deposition of pollutants to waterbodies and will continue to pursue implementation of programs available under various federal laws to reduce the human and environmental exposure to pollutants of concern.

(3) EPA is committed to supporting research activities that address the goals of CAA section 112(m).

In this chapter, Section V.A discusses EPA's assessment of the factors listed in CAA sections 112(m)(5)(A) through (E). This assessment is based on the scientific information summarized in this report, as well as the findings in the First Report to Congress. Section V.B discusses future directions and actions that EPA anticipates for the Great Waters program.

Section V.C summarizes EPA's draft determinations under section 112(m)(6), which are being published separately in the *Federal Register*. EPA also must consider under section 112(m)(6) two questions concerning the adequacy of the authority under provisions of section 112 and the possible need for further standards or control measures. Under the terms of a consent decree entered in *Sierra Club v. Browner*, Civ. No. 96-1680, EPA is to issue draft determinations for public notice and comment by June 30, 1997, and final determinations by March 15, 1998.

- ♦ EPA is required to determine whether the other provisions of section 112 are adequate to prevent serious adverse effects to public health and serious or widespread environmental effects associated with atmospheric deposition of hazardous air pollutants (HAPs) to the Great Lakes, the Chesapeake Bay, Lake Champlain, and coastal waters.

♦ Based on the information available in this report and on the determination regarding the adequacy of section 112 authority (which is specific to HAPs), EPA must also determine whether additional emissions standards or control measures under section 112(m)(6), beyond those otherwise authorized or required by section 112, are needed to prevent such effects from atmospheric deposition, including effects due to bioaccumulation and indirect exposure pathways.

V.A Reporting on the Role of Atmospheric Deposition to the Great Waters and Specific Actions Proposed

As stated in the CAA, EPA is required to assess the contribution of atmospheric deposition to the Great Waters on several issues (listed below). While these issues have been discussed at length at various places in the body of this report, this section summarizes EPA's efforts to address these five elements:

- Pollutant loadings — section 112(m)(5)(A);
- Environmental and public health effects — section 112(m)(5)(B);
- Pollutant sources — section 112(m)(5)(C);
- Exceedances of water quality or drinking water standards — section 112(m)(5)(D); and
- Description of any necessary revisions to requirements, standards, and limitations pursuant to the CAA and other federal laws — section 112(m)(5)(E).

Contribution of Atmospheric Deposition to Pollutant Loadings in the Great Waters

A substantial amount of scientific information has demonstrated that atmospheric deposition contributes to pollutant loadings in the Great Waters. In and around the Great Lakes, studies have shown that atmospheric transport and deposition of persistent hazardous chemicals occurs and is significant in the overall inputs to the lakes. The Great Waters program has focused on a set of chemicals that show persistence in the environment, tendency to accumulate in animal tissue, and toxicity to humans and other animals. Data from several studies of the lakes' waters, sediments, fish, and wildlife, show a general decrease in concentrations of persistent toxics during the 1970s and 1980s. These declines have resulted from many efforts taken to reduce discharges to both air and water of potentially harmful chemicals (which included the pollutants of concern). Also, some of these declines may be attributed to significantly reduced use or canceled registrations of persistent pesticides in the United States. In the 1990s, the overall picture of persistent toxics in the Great Lakes basin shows much more gradual decline, if any (though particulars vary from lake to lake and among pollutants). Due to limitations in historical monitoring techniques, considerable uncertainties exist for earlier data and thus, definitive conclusions about atmospheric pollutant trends are not made in this report. Quantitative monitoring data of atmospheric concentrations and deposition of the pollutants of concern have become available in recent years. It is anticipated that, as more data become available, better interpretations and conclusions can be made about pollutant trends, which in turn will better inform EPA about the extent to which additional actions will need to be taken, with possible focus on individual pollutants. Monitoring of toxics and nitrogen compounds (nitrates and others) has also been performed in Chesapeake Bay and other coastal estuaries and (with a focus on mercury) in Lake Champlain.

The Integrated Atmospheric Deposition Network (IADN) is an ongoing, binational monitoring network that assesses the magnitude and trends of atmospheric deposition to the Great Lakes region for wet and dry deposition and net gas exchange. Results from the past few years indicate that atmospheric deposition of some hazardous pollutants is still a concern for the Great Lakes. In similar work, data have been collected through the Chesapeake Bay Atmospheric Deposition Study (CBADS) on pollutant concentrations in air and precipitation. By incorporating emissions data with these measurements, atmospheric loading rates have been estimated. Certain trace metals appear to be deposited into the Bay as toxic contaminants. However, information on toxic contaminant trends in the Bay is not yet available. At Lake Champlain, researchers have been gathering information on deposition of mercury to the basin to characterize the types of mercury deposition (gaseous, wet, and dry particulate). To date, not enough information is available to assess trends for atmospheric mercury in the Lake Champlain basin.

Recent studies of several coastal waters have investigated the significance of atmospheric deposition to toxic pollutant loadings, and preliminary results show that atmospheric deposition can contribute a significant portion of loadings to coastal waters (e.g., Tampa Bay, Delaware Bay, Massachusetts Bays) for pollutants such as cadmium, lead, mercury, PAHs, and PCBs. For example, atmospheric deposition of cadmium is estimated to contribute 46 percent of the total annual load (direct and indirect) to Tampa Bay and 17 to 31 percent of the total annual load (direct) to Massachusetts Bays. Atmospheric deposition (direct and indirect) has been estimated to be a significant contributor of mercury (80 percent) to Delaware Bay. However, there are several remaining areas of uncertainty to be addressed related to atmospheric deposition of toxic contaminants to the Great Waters, including: the lack of accepted techniques for direct, quantitative measurement of dry particulate deposition and the unknowns associated with indirect loadings of pollutants, as they relate to watershed transmission and retention.

Information on relative loadings from air compared to other inputs of toxic contaminants in the Great Waters is still being assessed, although advances have been made in EPA's ability to address this issue. A comprehensive mass balance model for Lake Michigan has been initiated to gather information on loadings from different pathways, including the atmosphere, sediment, biota, and tributaries, as well as to study the variability in magnitude of atmospheric loadings near more urbanized areas versus rural areas. Preliminary modeling results from this major study are anticipated in 1998, and final results by 1999, which are expected to provide a better understanding of relative loadings and the movements of persistent pollutants in the environment.

Nitrogen deposition has been studied for coastal estuaries. Investigations have gathered information on atmospheric deposition estimates as well as relative loadings to these waters. Computer modeling studies for the Chesapeake Bay indicate that atmospheric deposition accounts for approximately 27 percent of the total annual loading of nitrogen (376 million pounds), while 23 percent of the load comes from point source water discharges and 50 percent from non-point sources other than air, such as fertilizers and animal wastes. Since the First Report to Congress, additional studies have been initiated to measure nitrogen deposition in coastal estuaries other than Chesapeake Bay (e.g., Tampa Bay, Long Island Sound, Massachusetts Bay). For those studies that evaluated both direct and indirect deposition, the contribution of atmospheric deposition to the nitrogen load ranges from approximately 12 to 44 percent.

To project the ultimate influence of changes in total nitrogen loadings to the Bay (including loadings other than from the air), the Chesapeake Bay Program recently configured the Bay Watershed Model to accept daily atmospheric loadings by land use category (i.e., forest, pasture, cropland, and urban). The Chesapeake Estuary Model is being upgraded to simulate basic ecosystem processes of submerged aquatic vegetation (SAV), benthic microorganisms, and major zooplankton groups. In addition, EPA's Regional Acid Deposition Model (RADM) is being directly linked to the Watershed and Estuary models. This new integrated model, functionally linking the airshed, watershed, estuary, and ecosystem, is expected to be completed in mid-1997. Some of the remaining areas of significant uncertainty in estimating atmospheric loads are: nitrogen retention in watersheds, the relative loadings of ammonia and organic nitrogen (compared to nitrate), and dry particulate deposition directly to water surfaces.

Contribution of Atmospheric Deposition to Adverse Human Health Effects or Adverse Environmental Effects in the Great Waters

Current information on potential exposure to and effects of pollutants of concern to the Great Waters adds greater weight to the scientific data and conclusions reached in the First Report to Congress. Effects on fish and wildlife that are associated with exposures to these persistent chemicals continue to be reported in research literature. At this time, it is not possible to identify which specific individual pollutants, or defined mixtures, cause specific adverse effects observed in the biota of the Great Waters. Even if more direct and definite links between exposure and effect were established, the relative contribution of a pollutant from current air deposition compared to overall loading for the waterbody still needs to be better characterized and quantified to provide a basis for setting appropriate reduction targets. Because many of the pollutants of concern persist for decades, current loading must be considered in light of contamination that has come from many sources and persisted many years. Persistence also means that while emissions and discharges of these chemicals may appear rather small in one year, the loading in the environment builds up over time. While pollutant compounds deposited from the air directly into a waterbody can have different routes of exposure to aquatic biota than the same pollutants brought in by water or in sediments, there are limited quantitative data on such differences in exposure. There is no current evidence that effects on wildlife, and potentially on human health, caused by specific pollutants deposited from the air are different from effects of those same chemicals carried in the water or sediment, once the chemical reaches an organism's tissues.

Exposure of wildlife or humans to the pollutants of concern can occur directly (e.g., through intake of drinking water, direct contact with water) or as a result of food web contamination. An indicator of potential human exposure to a pollutant is the presence of fish consumption advisories issued by state agencies. These waterbody-specific and fish species-specific advisories caution people against eating fish from a contaminated waterbody and suggest that consumption of fish with these levels of contamination may result in potential human health effects. According to EPA's 1995 national listing of fish and wildlife consumption advisories, current state advisories for the Great Waters are associated with the same pollutants of concern as in the First Report to Congress (see Appendix B). Fish consumption advisories in the lakes are most commonly issued for PCBs, followed by mercury and dioxins; in coastal waters, advisories are commonly issued for PCBs, followed by dioxins.

Periodic monitoring of pollutant levels in tissues of aquatic organisms, such as game fish, is another indicator of pollutant contamination and exposure and provides information on bioaccumulation in the food web. Most studies are based on measurements of pollutant concentration in fish from the Great Lakes. Studies of several pollutants show that, over many years, pollutant levels in fish are remaining constant or decreasing. In some instances, slight increases of some pollutants have been measured recently in certain fish species. For example, although significant declines in PCB and DDT concentrations in lake trout, walleye, and coho salmon in the Great Lakes were observed during the 1980s, more recently (i.e., in the last eight to ten years) the residues of PCBs and DDT in these fish have leveled off or even increased slightly. Chemical residues in fish tissue are probably the result of many factors in addition to deposition from the air. For example, the apparent increase in pollutants in fish tissue may be a result of resuspension of pollutants from sediment or from changes in the food web structure in the lake.

Adverse effects in wildlife and aquatic organisms in the Great Waters continue to be reported in the scientific literature. The correlations of tissue burdens of persistent pollutants with observed effects in the animals suggest that exposure levels for some pollutants continue to be high enough to produce adverse effects. Health problems persist for fish and other wildlife in certain Great Lakes locations, particularly for predators high in the food web, such as lake trout, mink, and bald eagles. Recent population studies of fish-eating birds in the Great Lakes (e.g., common terns) provide evidence of the developmental problems linked to PCBs during the 1980s.

The occurrence of health effects in humans is less studied in connection with general environmental exposures. Most information is based on laboratory studies in animals, which can provide useful insight on potential adverse effects in humans; however, for most chemicals, the lowest exposure concentration at which these effects would occur in humans is difficult to determine. The pollutants of concern have been investigated individually and are known to cause a variety of health effects, including cancer, and to act on many target organs, including the liver and kidney, and on the endocrine, reproductive, immune, and nervous systems. There are few recent studies of acute human exposures to Great Waters pollutants, although evaluations of people who frequently eat fish from the Great Lakes suggest concerns. The relative role of pollutants deposited from the atmosphere compared with other pathways contributing to exposure has not been quantified in these studies.

Although much information is available on potential exposure to the pollutants of concern and also on the potential effects of these pollutants, uncertainties still exist regarding the association between the actual exposure levels experienced over time and potential effects. Effects that occur in wildlife or humans from long-term exposure to pollutants of concern may differ from those effects caused by acute, high-level exposures because longer exposure to some chemicals can result in metabolism or breakdown of the chemical in the body to another chemical that is more or less toxic and can result in cumulative exposure due to bioaccumulation.

Comparison of the effects reported in laboratory studies to actual observations in the field (i.e., wildlife around the waterbodies) also needs to be addressed. Laboratory studies can provide evidence to support observations of effects in the field and, furthermore, give information on the possible mechanism of action of the effect. However, these studies generally do not duplicate the potential cumulative or combined effects that may occur when pollutants interact in the environment or simulate effects due to exposure in natural food chains.

Atmospheric deposition of nitrogen compounds can contribute significantly to eutrophication in coastal waters where plant productivity is usually limited by nitrogen availability. Eutrophication and its subsequent effects on estuarine ecosystems pose significant problems for Chesapeake Bay and many other coastal waters. Accelerated eutrophication frequently results in severe ecological effects such as nuisance algal blooms and reduced oxygen levels in the water (as unstable algal blooms die back or organisms sink into deeper water). The reduction in oxygen levels may result in altered food webs by reducing or eliminating bottom-feeder populations of fish or shellfish, creating conditions that favor different species, or causing dramatic fish kills. In some cases, overproduction of algae increases the suspended matter in the water, which decreases light penetration to submerged aquatic vegetation (SAV) or coral. In other cases, algae can overgrow directly on submerged living organisms, often with losses of SAV or coral communities. Major areas of uncertainty include the response of living resources, particularly SAV, to reductions in nitrogen loads and the tidal-flow exchanges between coastal estuaries and offshore waters, which also receive atmospheric deposition of nitrogen compounds. Further study is needed on the direct link between atmospheric deposition of nitrogen compounds and ecosystem responses. Oxides of nitrogen are important components in acidic deposition, wet and dry, to fresh waters. In addition, nitrogen compounds and their reaction products in the ambient air can produce direct impacts on human health and terrestrial vegetation. In consideration of these partial impacts, EPA has established programs to evaluate and reduce the threat to human and ecological health from atmospheric nitrogen oxides, ozone, particulates, and acidic aerosols.

Emission Sources that Contribute to Atmospheric Deposition in the Great Waters

For toxic contaminants, information linking specific emission sources to impacts on atmospheric deposition to the Great Waters has been limited. Some work has been done on identification and characterization of specific sources in the Great Lakes, and more is planned. Some of this work does not involve emission inventories, but consists of predicting the origin of an air mass (using meteorology) that passes over a monitoring station and determining emission sources based on observed pollutants and metals ratios in the air samples. For some source categories, there currently are inadequate reliable emissions data and detailed emissions inventories to link pollutant deposition to specific sources. However, for many source categories, substantial progress has been made in recent years to establish source emissions inventories through state and federal efforts. For example, the Great Lakes Emissions Inventory, recently implemented by the eight Great Lakes states and the Canadian Province of Ontario, is compiling a data base on emission sources of 49 toxic air pollutants that will provide information on local and regional sources of pollutants of concern. A pilot project of major urban areas along the southwest shore of Lake Michigan has created an inventory of small point and area source categories in the surrounding region that contribute the most to the total emissions of major pollutants. As a result of this pilot study, a better methodology has been developed for use in the full regional Great Lakes Emissions Inventory. Research studies on the importance of local "urban plumes" of pollutants have been underway in recent years near Chicago (impacting Lake Michigan) and near Baltimore (impacting Chesapeake Bay). These studies are due to be completed in the next few years. Their results, combined with emission inventories, will assist in defining the relative importance of local emissions upon deposition and in focusing emission reduction efforts. In addition, EPA recently completed a national emissions inventory of known U.S. sources of seven HAPs listed under CAA section 112(c)(6). Identification of the sources for total emissions of these pollutants is necessary to assure that at least 90 percent of emissions from stationary, anthropogenic sources are subject to regulation under CAA section 112(d).

As a result of research funded by the Great Waters program and others, better predictions can be made on the direction and movement of emitted air pollutants over large geographic areas, and more reliable estimates of rates at which pollutants will be deposited on land or water surfaces are possible. For example, some researchers are determining the extent of mercury emissions to air in the United States over an entire year, the deposition to the land and waterbodies, and the contribution by source category to the total amount of mercury emitted and deposited within the United States. In the Great Lakes, a regional network of ten monitoring sites was established in 1993 by EPA and the University of Michigan to measure atmospheric mercury and wet and dry deposition of gases and particulates. One goal is to determine the sources and source areas of mercury deposition to the Basin, using an improved trajectory clustering technique. Such studies will enable EPA and other researchers to better explore source attribution.

What is CAA Section 112(c)(6)?

Under this section, EPA is required to identify and list sources, categories, and subcategories of alkylated lead compounds, hexachlorobenzene, mercury, PCBs, POM, TCDD, and TCDF. This list is intended to assure that not less than 90 percent of the aggregate emissions of each pollutant are subject to standards under section 112(d)(2) or 112(d)(4). To meet these requirements, national inventories of sources and emissions of these pollutants have been developed. Because all seven pollutants are of particular interest to the Great Waters program, these inventories will be useful in protecting the Great Waters and in implementing section 112(m).

Substantial progress also has been made on investigating emission sources of nitrogen compounds in recent years. Due to the impacts that nitrogen overenrichment has in estuaries and coastal waters, sources of airborne and waterborne nitrogen compounds are being evaluated by monitoring and modeling studies. Recent modeling studies have looked at both local and distant sources (generic types) that release atmospheric nitrogen that deposits into the Chesapeake Bay and its watershed. Using RADM, the Chesapeake Bay Program has evaluated sources contributing to the deposition of certain nitrogen compounds and identified electric utilities and mobile sources as major contributors. However, the model runs show that the patterns of nitrate deposition are different for these two broad source categories. Utilities appear to contribute a majority of the nitrate that deposits on the western side of the Bay watershed and exhibit a decreasing trend in deposition from the western towards the eastern portion of the watershed. Mobile source emissions, on the other hand, largely reflect the traffic associated with the Washington, D.C./Boston corridor and contribute the majority of the nitrate that deposits along the Delmarva Peninsula, the Bay itself, and lower portions of the western shore tidal tributaries. Work on addressing a modeling uncertainty in RADM — correct partitioning between particulate nitrate and nitric acid — continues with funding from EPA's High

Performance Computing and Communications Program, the Particulate Program, and the Acid Rain Program.

Contribution of Atmospheric Pollutant Loading to Exceedances of Water Quality Standards and Drinking Water Standards or Exceedances of Objectives of the Great Lakes Water Quality Agreement

While some pollutants remain at levels exceeding applicable criteria in some locations of the Great Waters, no additional pollutants have been found to exceed water quality standards in the Great Waters since the First Report to Congress. Where exceedances occur, it may be reasonable to assume that atmospheric deposition contributes to some (quantity unknown) extent. The contribution of atmospheric deposition is especially likely for open water exceedances, in contrast to localized "hot spots" where other loading mechanisms may dominate. Where data are available, toxic contamination in water does not appear to be increasing significantly for any of the pollutants of concern, but rather appears to be remaining the same or decreasing. However, recent data on water column concentrations are available for only a limited number of the pollutants. Concentrations of DDT/DDE, dieldrin, hexachlorobenzene, and PCBs in the water column of the Great Lakes are reported in both this report and the First Report to Congress. Current levels of dieldrin and PCBs are lower than those presented in the First Report to Congress; however, they continue to exceed the most stringent water quality criteria for the Great Lakes by a substantial margin (tens to hundreds of times). These most stringent criteria incorporate biomagnification up the food chain, and so differ from criteria for direct exposure to water. In addition, the reported concentrations of DDT/DDE may exceed the current water quality criteria under the Great Lakes Initiative; however, the possible exceedance is difficult to establish given analysis limitations. Thus, even though mass emissions of these pollutants have decreased dramatically as a result of actions taken to ban or restrict the manufacture and use of these substances, the long-term persistence of these pollutants in the environment, and apparent cycling among media, cause these compounds to remain pollutants of concern to the Great Lakes. Therefore, these pollutants warrant continued monitoring and tracking until levels no longer exceed relevant criteria or standards.

Among the Great Waters, the only significant sources of drinking water are the Great Lakes and Lake Champlain. The drinking water systems using these sources are not known to have exceedances of drinking water standards for the pollutants of concern, including toxics and nitrates.

Description of Revisions to Requirements, Standards, or Limitations Pursuant to the Clean Air Act and Other Applicable Federal Laws, as Necessary

Section 112(m)(5)(E) requires that EPA describe in this report any revisions to the requirements, standards, and limitations pursuant to the Clean Air Act and other applicable federal laws that are necessary to assure protection of human health and the environment. In reviewing relevant requirements, standards, and limitations established pursuant to applicable statutes, EPA has found that such restrictions are achieving significant reductions in releases of the pollutants of concern. For example, the technology-based standards that have been, and are being, developed and promulgated for stationary air sources require reductions in air emissions that reflect the maximum that are achievable, in accordance with section 112(d). Section 112(c)(6) specifically lists for special attention seven pollutants: alkylated lead compounds, polycyclic organic matter (POM), hexachlorobenzene, mercury, PCBs, furans (2,3,7,8 TCDF), and dioxins (2,3,7,8 TCDD). These seven pollutants are also Great Waters pollutants of concern. A comprehensive emission inventory has been developed under section 112(c)(6), and further standards are being developed under section 112(d).

As mentioned earlier, section 112 authority is specific to HAPs listed under section 112(b) and, therefore, does not apply to pollutants that are not currently listed, such as nitrogen compounds. However, several major programs have been implemented under other sections of the Clean Air Act to reduce emissions of oxides of nitrogen: sections 108 and 109, pertaining to ambient air standards for stationary sources to protect public health and welfare; section 202, pertaining to air regulations for mobile sources; and section 407, pertaining to acid deposition control.

Nitrogen dioxide is listed under section 108 as a pollutant that causes or contributes to air pollution that may reasonably be anticipated to endanger public health or welfare. Air quality criteria have been issued by EPA for nitrogen dioxide, and ambient air quality standards have been established for nitrogen dioxide under section 109, which directs the EPA Administrator to propose and promulgate primary and secondary National Ambient Air Quality Standards (NAAQS) for pollutants identified under section 108. A primary standard is one that is necessary to protect the public health, allowing an adequate margin of safety. A secondary standard, as defined in section 109, must specify a level of air quality needed to protect the public welfare from any known or anticipated adverse effects associated with the presence of the pollutant in the ambient air. Welfare effects, defined in section 302(h), include, but are not limited to, effects on soils, water, crops, vegetation, materials, animals, wildlife, weather, visibility, and climate. Thus, section 109 provides authority to address a broad range of public health impacts and adverse environmental effects in order to assure protection of human health and the environment.

Several efforts are currently underway to control emissions of nitrogen oxides. The Ozone Transport Assessment Group, a regional body representing 38 eastern and midwestern states, is currently evaluating alternative reduction options for regional control of nitrogen oxides and volatile organic compounds in order to achieve attainment of the current NAAQS for ozone. In addition, EPA is considering revisions to the NAAQS to address the most recent information about health and environmental effects of ozone and fine particulate matter. A subcommittee of the Clean Air Act Advisory Committee chartered under the Federal Advisory Committee Act is evaluating regional approaches to the management of emissions of nitrogen oxides, sulfur dioxide, fine particulate matter, and volatile organic compounds to attain the proposed revisions to the ozone and particulate matter NAAQS and the national goals of the regional haze program. Together these efforts are expected to result in substantial reductions in emissions of the pollutants of concern, with significant additional benefits to geographic areas such as the Chesapeake Bay watershed.

Furthermore, section 202 requires that standards for motor vehicles be established, as needed, to reduce emissions that cause or contribute to air pollution that may reasonably be anticipated to endanger public health or welfare. Regulations under this section apply to emissions of nitrogen oxides. Section 401 (Acid Deposition Control) identifies the presence of acidic compounds and their precursors in the atmosphere, and in deposition from the atmosphere, as a threat to natural resources, ecosystems, materials, visibility, and public health. The principal sources of acidic compounds and their precursors in the atmosphere, as identified in section 407, include nitrogen oxides from the combustion of fossil fuels. A program for reducing emissions of nitrogen oxides from coal-fired electric utility power plants has been promulgated under section 407 of the Clean Air Act. In the Acid Rain Phase II NO_x Emission Reduction Rule (*Federal Register*, December 19, 1996), water quality benefits are cited in the preamble justification as a basis for EPA's decision to exercise statutory discretion to lower emission limits for certain coal-fired utility boilers (known as Group 1) and as an environmental impact of establishing emission limits for other boiler types (known as Group 2). Eutrophication is listed with ozone and acid deposition in the summary table of environmental effects.

Pesticides that are included as pollutants of concern have been, and continue to be, effectively addressed by actions taken under relevant statutes. Also, PCB releases have been, and are being, effectively reduced. While concentrations of some of these pollutants of concern in the waterbodies or in animal tissue remain unacceptably high, current releases of these pollutants are being effectively managed. It is important to note that, although current efforts are effective and appropriate, some level of risk to the public health and to the environment may continue for years into the future due to previous releases of the pollutants of concern and their persistence in environmental media for multiple decades.

Various efforts are being taken by EPA that will provide significant concomitant benefits through reduction of pollutants EPA addresses under section 112(m). At this time, no specific revisions to requirements, standards, and limitations pursuant to the Clean Air Act or other relevant federal statutes have been identified as necessary to assure protection of human health and the environment in response to section 112(m) assessments. In the future, as EPA evaluates progress of ongoing efforts and considers new information as it becomes available, new approaches may be pursued.

V.B Future Directions

As described throughout the report, notable progress has been made to further the knowledge of atmospheric deposition of pollutants to the Great Waters. As new information becomes available on atmospheric pollutant deposition to the Great Waters, questions or issues are expected to arise that will require further investigation or action. At this time, EPA has identified areas where information is limited and has identified some specific directions that need to be taken to advance the understanding of issues relevant to the Great Waters program.

Determine Management/Regulatory Actions for Focus Pollutants

EPA plans to continue evaluating the 15 pollutants of concern, focusing on those that are currently being emitted to the air from sources subject to regulations under the CAA. For example, emphasis will be placed on the seven pollutants of concern addressed in section 112(c)(6). Emission inventories have been developed for these pollutants and will provide useful information on sources of atmospheric deposition to the Great Waters. EPA's Air Toxics program has recently prepared some notable studies that may have implications to the Great Waters pollutants of concern, including the Interim Report to Congress on emissions of hazardous air pollutants from electric utilities. The Interim Report, along with the Great Waters Second Report to Congress and sponsored studies, contribute information focused on persistent pollutants to EPA's ongoing actions establishing emission standards under section 112(d). As these standards continue to become implemented, significant reductions in HAPs are expected, including the persistent chemicals of importance to waterbodies. In addition to these activities, EPA is considering future management and/or regulatory actions for some pollutants of concern, some of which are described below.

Continue Monitoring and Research Efforts to Support Management/Regulatory Actions

EPA plans to continue supporting monitoring and research efforts that provide information for regulatory and management actions for pollutants of concern. Monitoring allows tracking of current and future reductions and evaluation of EPA program effectiveness. In addition, monitoring supports the development and validation of atmospheric transport models that enhance EPA's program implementation and predictive capabilities. Research projects include work that expands current monitoring capabilities, increases knowledge of cycling and transport of contaminants, and evaluates environmental effects.

♦ ***Exposure and Effects:*** A process to coordinate research strategies on persistent pollutants has begun among several offices within EPA. It is expected that these studies will improve our understanding of the relationship between ambient concentrations in natural media (water, air, and sediments), burdens of pollutants in tissues of living biota, and associated effects. Exposure and effects research will build on methods used to develop the Great Lakes Water Quality Criteria, which incorporate consideration of biomagnification. These types of research are central to increasing our ability to assess the impact of atmospheric deposition to the Great Waters and define efficient approaches to reducing exposures and risks to humans and wildlife.

♦ ***Mercury and Compounds:*** Mercury is a global pollutant, and is used in industries and released from combustion, manufacturing, and natural processes. Through changes in its ionic and molecular chemistry, mercury can be mobile among the atmosphere, waters, biota, and soil or sediments. EPA is committed to identifying feasible options for reducing or preventing emissions of mercury. Also, following completion of EPA's Mercury Study Report to Congress, the results will be used in the process of identifying sources of mercury emissions and in prioritizing mercury reduction strategies. To date, most routine monitoring for atmospheric mercury does not provide key information needed to evaluate the sources and impacts of mercury loadings. This is due to the relatively recent development of mercury techniques that differentiate between the various molecular states of mercury and have not yet been widely and routinely used. However, these techniques have been used on a research basis concurrent with monitoring for other metals, in order to obtain an atmospheric "fingerprint" that aids in the identification of sources. Monitoring for mercury is expected to continue in biota, atmosphere, and other components of ecosystems, particularly in the Great Lakes region. Monitoring should continue, to track future reductions of mercury, as emissions in the United States are expected to decrease upon implementing recent CAA regulations such as the municipal waste combustor rule, and the proposed standards for medical waste incinerators and for hazardous waste combustors. The effectiveness of voluntary initiatives also should be evaluated.

- ***Combustion Emissions:*** Combustion emissions (POM/PAHs, dioxins, furans, and PCBs) are part of the EPA's toxics control efforts utilizing regulation, pollution prevention, voluntary measures, and other approaches. Releases that affect the Great Waters include past usage, combustion (including combustion of wastes), and cycling or movements among the components of ecosystems. For example, although PCBs have not been manufactured for a long time, they remain in use within existing electrical equipment. When released into the environment, PCBs have proven to be one of the most persistent of the pollutants of concern. Regulatory action to control these pollutants under the CAA can be complex because current emissions of these toxics from any one source may be a small percent of total loadings. However, their persistence and the tendency of many of the chemical species to biomagnify gives them a particular importance. Research on effects to wildlife associated with environmental exposures, including mixtures of toxics, is expected to continue. Monitoring of fish-eating birds and other wildlife in the Great Lakes should be performed with respect to their geographic relation with the Areas of Concern (AOCs). The compounds grouped under POM, including PAHs, have had less attention in monitoring in the past, but because they are still being emitted, special focus on monitoring and studies of effects are expected. PAH trends are currently tracked in the Great Lakes.

- ***Pesticides:*** Persistent pesticides (chlordane, DDT/DDE, dieldrin, hexachlorobenzene, α-HCH, lindane, and toxaphene) are difficult to control through regulation under the CAA since the most significant potential sources to the Great Waters may not be from current emissions. Releases by several pathways to the Great Waters are due in many cases to past use, often over considerable areas, while cycling or movements among Great Waters' ecosystem components can release earlier contamination. In addition, current human activities, including combustion, can still emit small quantities. Use of all the pesticides of concern are either canceled or severely restricted in the United States, yet they continue to be found in sediments of waterbodies. Research is expected to focus on possible current sources as byproducts or long-range transport of toxaphene and dieldrin due to their continued importance in some of the Great Lakes. These chemicals are retained on the list of Great Waters pollutants of concern, and EPA believes that efforts at preventing and controlling releases of these chemicals are warranted, including continued monitoring until levels no longer warrant fish advisories or exceed water quality criteria and standards.

- ***Nitrogen Compounds:*** EPA is focusing on management strategies to reduce nitrogen oxide (NO_x) emissions. EPA expects that total NO_x emissions will gradually decline about six percent from current levels by the year 2000 due to mandatory CAA programs. After the year 2000, the total national NO_x emissions are expected to gradually increase due to population growth. EPA is developing the Integrated NO_x Strategy, which outlines an integrated approach to controls on mobile and stationary sources through cost-effective mechanisms. The Strategy stresses that consideration of the many environmental effects of NO_x emissions should strongly influence policy decisions regarding the control of NO_x emissions. Accordingly, EPA continues to work in coordination with a wide range of stakeholders to develop and implement new mobile and stationary source control programs at the federal, state, and local levels to reduce NO_x emissions. Monitoring of future reductions in conjunction with estuarine, watershed, and acid deposition studies is expected to continue. Monitoring should continue to include a comprehensive approach that addresses wet and dry deposition of gases and particulates. Additional research will proceed on improving monitoring methods for dry particles and on quantifying the relative contribution of the chemical species of nitrogen compounds to total loading. Modeling will be important in addressing the multiple questions and benefits that strategies for reducing NO_x are developing in conjunction with programs on ozone and fine particulate matter, as well as impacts on waterbodies.

Expand Modeling Efforts to Estimate Atmospheric Loadings to Great Waters

As data continue to be gathered from studies that were initiated in recent years, the results will lead to better characterization of atmospheric loadings to the Great Waters. One particular study that should provide comprehensive information is the Lake Michigan Mass Balance Model. This model should be adapted and applied to additional waterbodies. EPA also believes that more investigation is needed to assess the contribution of atmospheric deposition to pollutant loadings from urban, stormwater, and agricultural runoff from the watershed. This kind of work is currently being performed for nitrogen compounds in Chesapeake Bay. Results from such projects will provide more comprehensive information to assess the complete picture of atmospheric loadings to the Great Waters.

Increase Focus on Identification of Emission Sources

Significant progress has been made in establishing source emission inventories for some pollutants of concern. However, information is limited on specific sources of atmospheric deposition to the Great Waters, especially sources that are relatively distant from the waterbody. Because this information is critical to developing risk management strategies, EPA believes that more effort should be placed on determining potential sources of these airborne pollutants, with emphasis on identifying significant sources, both local and long range. As described above, source emission inventories have been initiated in the Great Lakes region and nationally, which will be important in identifying source categories of concern. The Chesapeake Bay Program has made important progress in identifying local and distant sources for nitrogen deposition to the basin, but has not determined the specific sources that contribute the most to deposition.

Another future area of study for EPA is atmospheric pollutant deposition from sources other than current air emissions. As discussed earlier, many of the pollutants of concern to the Great Waters are no longer used or manufactured in the United States; however, they cycle in the environment, may be transported considerable distances, and thus can contribute some unknown amount to the waters. Investigations on the relative contribution of deposition from current activities versus past use is an important concern to EPA because several of these pollutants may need to be addressed through approaches that supplement prevention measures and emission controls developed under the CAA.

Continue to Promote Pollution Reduction in the Great Waters

In the past few years, local, regional, and federal agencies, as well as international organizations, have worked together to develop initiatives and agreements that promote activities supporting pollution prevention and reduction efforts in the Great Waters. As such, several of these activities will help to address issues under CAA section 112. Many of these programs, which are described in Chapter IV of this report, were developed recently, and implementation of these initiatives is expected to occur in the near future. EPA is committed to continue its support for, and involvement in, these activities. Some of these broad initiatives are highlighted below:

- ***Strategic pollution prevention plans developed through federal, state, and local partnerships.*** Many waterbodies of the Great Waters currently have specific goals or strategies to characterize and/or reduce water pollution in their respective ecosystems. Most plans promote comprehensive approaches to reduce contamination in a waterbody. Where atmospheric deposition is a significant contributor, EPA is committed to using its section 112 authorities to assist in achieving the emission reductions feasible. As appropriate, other authorities may be considered as well (e.g., provisions of the CAA to reduce emissions of nitrogen compounds).

- ***Use of voluntary efforts to meet pollution reduction goals.*** Pollution reduction goals have been developed for many Great Waters and, in most instances, implementation of these goals involves voluntary initiatives. For example, state agencies and other stakeholders in the Great Lakes region have worked together to propose voluntary, as well as regulatory, measures to prevent or reduce atmospheric mercury contamination in the Great Lakes through increased public awareness, development of alternative technologies, and capturing and recycling of uncontrolled (fugitive) releases. Many activities by industries, individuals, and cities that reduce waste of energy and develop efficient transportation also have the benefit of reducing NO_x emissions.

- ***Binational and international efforts to promote pollution reduction.*** Binational efforts can play a critical role in controlling atmospheric pollution in the Great Lakes. For example, the Great Lakes Water Quality Agreement and the recently signed Binational Strategy for Virtual Elimination between the United States and Canada have set percentage reduction goals as steps towards virtual elimination of persistent, toxic, and bioaccumulative pollutants from the Great Lakes. Through these endeavors, both nations are encouraging and supporting voluntary programs and other actions to reduce generation, use, and release of toxic contaminants to the Great Lakes. In addition, EPA works in cooperation with other agencies and departments to develop and support additional binational and global actions to address atmospheric sources of persistent organic and heavy metal emissions.

- ***Implementation of the Great Lakes Water Quality Initiative.*** The primary purpose of the Great Lakes Water Quality Guidance (GLWQG) is to provide a consistent level of protection to people, aquatic life, and wildlife that may be exposed to toxic pollutants from the Great Lakes. To accomplish this goal, the GLWQG establishes protective levels, or water quality criteria, for toxic pollutants from all sources. Estimates of basin-wide toxic reductions that will result from implementation of the Great Lakes Water Quality Initiative range from 2.6 million to 3.5 million kilograms of toxic pollutants per year.

- ***Continue to take strategic actions.*** Some argue that any continuing releases of persistent bioaccumulative pollutants add to an environmental burden that is already causing effects. EPA believes it is important to balance its present understanding of atmospheric deposition against the implications of inaction in order to define those actions that are justified at this time. EPA is committed to protecting public health and the environment and will promote taking whatever regulatory actions and voluntary initiatives are appropriate in the most cost-effective way possible.

Assess Economic Impact of Pollution to the Great Waters

The economic impacts associated with reductions of pollutants to the Great Waters have not been sufficiently investigated, although a regulatory impact analysis was recently performed on the Great Lakes Water Quality Guidance, which addressed the impacts of the new guidance. For the Great Waters program, EPA plans to identify and quantify, where possible, economic impacts associated with exposure and effects indicators such as fish advisories, habitat decline, diminished species diversity, fish kills, and declining populations of contaminated shellfish and fish.

V.C Draft Determination of Whether CAA Section 112 Authorities are Adequate to Prevent Adverse Effects to Public Health and the Environment from Deposition of HAPs

In accordance with section 112(m)(6) of the CAA, EPA is issuing, at the same time it submits this Report to Congress, a draft determination of the adequacy of the other legal authorities and mandates provided by section 112 of the CAA to prevent specified adverse human health and environmental effects associated with atmospheric deposition to the Great Waters. Based on the information available in this report and in the draft determination regarding the adequacy of section 112 authority, EPA is also issuing a draft determination of whether additional emissions standards or control measures, beyond those authorized or required by section 112, are needed to prevent such effects. These draft determinations are described in a *Federal Register* notice published separately and are briefly summarized below.

Section 112(m)(6) of the CAA requires that EPA determine whether adequate authority exists within the other (i.e., non-Great Waters) provisions of section 112 to prevent serious adverse effects to public health and serious or widespread environmental effects resulting from atmospheric deposition of HAPs to the Great Waters. In making this determination, EPA reviewed the authority granted by the other provisions, as they may apply to deposition of HAPs to the Great Waters. It should be emphasized that this determination pertains to the authority within the CAA to take actions as appropriate to address adverse effects.

In addition, EPA has focused on the authority within section 112 to address those pollutants and sources within the scope of section 112. Therefore, pollutants such as nitrogen compounds that are not on the section 112(b) list of HAPs are not within the scope of this determination. Similarly, sources that are regulated by other sections of the CAA (e.g., mobile sources) or that are addressed by other statutes (e.g., wastewater discharges, which are addressed by the Clean Water Act) are also not within the scope of this adequacy determination.

Section 112 establishes a statutory scheme by which EPA is to identify HAPs that may cause or contribute to adverse effects to public health or the environment, develop standards for the control of emissions from stationary sources of such HAPs, and adjust these control requirements as needed to address any remaining unacceptable risk that may be present after imposition of sources have complied with the emission standards. The types of adverse environmental effects to be prevented are defined in the Act and are broad in scope.

Authorities provided by other provisions of section 112 that may be particularly relevant to the Great Waters pollutants and sources are briefly summarized below and, as stated earlier, are described in more detail in the *Federal Register* notice. Section 112 authorizes EPA to:

- Identify and list additional air pollutants that may cause adverse effects due to atmospheric deposition (section 112(b));
- Identify and list any stationary source category that emits pollutants with the potential to cause adverse effects (section 112(c));
- Establish a lesser quantity (e.g., below ten tons per year for a single pollutant) emission rate for defining major sources based on several factors, including persistence and potential to bioaccumulate (section 112 (a)(1));
- Promulgate performance standards based on best performing technologies for major sources and listed area sources (section 112(d)); and
- Require additional controls, beyond the section 112(d) standards, as necessary to provide an ample margin of safety to protect public health or to prevent an adverse environmental effect (section 112(f)).

Based on its analysis of these and other section 112 provisions, EPA has issued a draft determination that section 112 authority is adequate to prevent serious adverse effects to public health and serious or widespread environmental effects associated with the deposition of HAPs to Great Waters. Consequently, EPA also has issued a draft determination that, at this time, no further emissions standards or control measures beyond those authorized under the other provisions of section 112 are necessary and appropriate for stationary sources of HAPs to prevent such effects. EPA has requested public comment on these draft determinations, which will be finalized in March 1998.

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APPENDIX A
STATUS OF ACTIONS RECOMMENDED IN FIRST REPORT TO CONGRESS

This appendix provides information on the status of the actions that were recommended in the First Report to Congress on atmospheric deposition to the Great Waters (see Table A-1) and lists the emissions standards that may control the Great Waters pollutants of concern (see Table A-2).

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
1. EPA will continue ongoing efforts to implement section 112 and other sections of the Clean Air Act, as amended in 1990, and will use the results of this report in taking reasonable actions to reduce emissions of Great Waters pollutants of concern.	
a. EPA is developing standards under section 112(d) for approximately 35 source categories of Great Waters hazardous air pollutants (HAPs) of concern, consistent with the schedule published in response to section 112(e)(3). Where possible, given other factors, EPA will publish section 112(d) standards ahead of schedule for specific source categories. Great Waters Program funds will be used to develop and publish ahead of schedule section 112(d) standards for at least one source category.	Fiscal year 1994 Great Waters funds supplemented the development of the primary aluminum maximum achievable control technology (MACT) regulation. As a result, the standard was proposed on September 26, 1996. This regulation has been developed via a successful partnership with industry, states, and environmental and tribal interests. The standard primarily addresses fluoride emissions but will also include limits for polycyclic organic matter (POM), the Great Waters pollutant that made this source category a choice for Great Waters funding. Promulgation of this standard is expected in late 1997.
b. During the process of developing emission standards, EPA will evaluate whether the currently defined MACT floor for existing sources represents a sufficient level of control for sources that emit Great Waters pollutants of concern.	At present, the tools to quantitatively evaluate a "sufficient level of control" for Great Waters pollutants do not exist. Nonetheless, in support of this recommendation, Great Waters pollutants are being considered as various source categories are evaluated for pollution control. Table A-2 lists emission standards currently being developed, or which have been completed, that address emissions of some of the Great Waters pollutants of concern.
c. As soon as practicable, EPA will publish an advance notice of proposed rulemaking (ANPR) to notify the public of EPA's interest in establishing lesser-quantity emission rates or LQERs (i.e., less than 10 tons per year) for selected Great Waters HAPs for the purpose of defining sources emitting these HAPs as "major sources" and to solicit comment. EPA will also evaluate whether any Great Waters HAPs warrant establishment of an LQER, and, if appropriate, based on that evaluation and the comments on the ANPR, EPA will develop a notice that proposes LQERs for those pollutants for which an LQER is warranted.	At this time, EPA has postponed development of LQERs. The primary impact of developing an LQER for a pollutant or source category is the consequent definition of that source as a "major" source and thus subjecting that source to certain requirements under CAA section 112. Most sources of the pollutants for which LQERs were being considered are already defined as major, and thus the establishment of an LQER would have little effect. For those sources that are area sources, performing an "area source finding" is a more efficient way to assess source categories as they come up for regulation rather than an up-front LQER analysis with more generic data. Also, there is sufficient statutory authority to require MACT on sources of any size regardless of their definition as major or area sources. In the future, however, EPA may decide that LQERs are warranted for specific source categories or pollutants at which time this effort could be re-initiated.

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
d. During the process of standards development for major sources, EPA will determine whether area sources of Great Waters HAPs warrant regulation under section 112(d) and, if so, which area sources. Results of the assessment will be integrated into the strategy for area sources under development in accordance with section 112(k).	EPA has evaluated 15 area source categories to determine whether regulation of these sources is warranted under section 112(d). These included four source categories emitting lead or dioxins. To date, none of the source categories emitting these Great Waters pollutants has warranted an "area source finding" based on a risk assessment. Area source analyses will continue to be performed as appropriate.
e. For the urban area source strategy (section 112(k)), EPA will evaluate public health effects on the basis of total exposure, which would include exposure by inhalation as well as exposure through ingestion of food containing bioaccumulated urban toxicants.	To assess total exposure to HAPs and criteria pollutants, EPA is currently developing the Total Risk Integrated Model (TRIM). The TRIM will be a probabilistic model capable of assessing risks to humans and to populations in an ecosystem resulting from multimedia contamination (in air, water, soil, and food) and multipathway exposure (via inhalation, ingestion, and absorption exposure routes). The TRIM is expected to be available for use by December 1999.

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
f. EPA will conduct a pilot project examining the use of Great Waters impacts analyses in the development of section 112(d) standards.	EPA has recently developed a new modeling tool for the assessment of atmospheric deposition of pollutants to the Great Waters. REMSAD, the Regulatory Modeling System for Aerosols and Deposition, is a work station-based Eulerian model intended for use in assessing the impacts of regulatory activities, such as section 112(d) MACT standards, on loadings of pollutants of concern to the Great Waters. REMSAD is currently capable of simulating short-, medium- and long-range transport and deposition of mercury, cadmium, dioxins, and POM; other pollutants, including other toxics as well as nitrogen, may be incorporated in future work. Initial model demonstration and evaluation will be completed during 1997. The model is currently available on the OAQPS Support Center for Regulatory Air Models (SCRAM) bulletin board. As part of the REMSAD development and demonstration activities, EPA will conduct a pilot study that will examine the impacts of the Emissions Guidelines for Municipal Waste Combustors (MWCs) on deposition of pollutants of concern to the Great Waters. Like section 112(d) standards, the MWC standard is based on MACT. The MWC category was chosen because it satisfied several requirements for an effective pilot study: (1) it includes a large number of sources; (2) it emits significant quantities of several pollutants of concern, including dioxins, furans, cadmium, mercury, and POM; (3) and suitable emissions data are available. The pilot study will estimate changes in annual loadings of these pollutants to the Great Waters due to implementation of the MWC standard.
g. For Great Waters HAPs, EPA is proposing a cap (i.e., 0.01 ton per year) to the de minimis levels being developed under section 112(g), so that controls would be required for more sources of Great Waters HAPs as they modify their processes. EPA will determine the appropriate de minimis level on a chemical-by-chemical basis, giving consideration to the chemical's persistence, propensity to bioaccumulate, and such other factors that EPA considers relevant.	In the proposed section 112(g) rulemaking, de minimis emission rates were proposed for HAPs identified as being of concern for Great Waters (based on toxicity, bioaccumulation, and bioconcentration). However, the 112(g) rulemaking that was promulgated did not include the provisions pertaining to modifications and, thus, the de minimis levels were not needed and were not included in the final rule.

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
h. EPA plans to propose a revised MWC rule, with stringent controls on mercury emissions and emissions of other Great Waters HAPs, not later than summer 1994.	On September 20, 1994, EPA proposed New Source Performance Standards (NSPS) and Emission Guidelines (EG) applicable to MWC plants larger than 35 megagram (Mg) per day capacity. These regulations were finalized on October 31, 1995. For mercury, the final standard for new and existing MWCs is 0.08 milligram (mg) per dry standard cubic meter (dscm), or about 90 percent control. The final rules also apply to dioxins. The air pollution control system used to comply with the CAA section 129 guidelines achieves greater than 95 percent dioxin reduction.
i. EPA is conducting studies that will provide information for future Great Waters reports. The mercury study, under section 112(n)(1)(B), will evaluate the rate and mass of mercury emissions from all sources, the health and environmental effects of such emissions, technologies to control such emissions, and the costs of these control technologies. The utility study, under section 112(n)(1)(A), will evaluate the hazards to public health reasonably anticipated to occur as a result of emissions of all HAPs by electric utility steam-generating units. Findings of these studies will be relied upon in future Great Waters reports in the development of strategies for reducing environmental exposures to Great Waters pollutants.	EPA has prepared a draft seven volume Report to Congress on mercury which was submitted to EPA's Science Advisory Board for peer review in June 1996. The report was favorably reviewed in February 1997 and EPA expects to finalize and submit the Report to Congress in fiscal year 1998. In October 1996, EPA submitted to Congress an interim report on utility air toxics (U.S. EPA 1996e). Entitled *Study of Hazardous Air Pollutant Emissions for Electric Utility Steam Generating Units -- Interim Final Report* (EPA-453/R-96-103abc), this document addresses inhalation and non-inhalation exposures to utility emissions. A final report, including a regulatory determination for utility control under section 112 is due January 15, 1998. The Executive Summary of the interim Utility Study can be accessed through EPA's Technology Transfer Network (TTN) by calling (via modem) 919-541-5742.
j. EPA is developing ecological effects assessment screening methods for reviewing petitions to add and delete pollutants from the HAP list and to delete source categories from the source category list. EPA will consider the Bioaccumulation Factor Methodology (58 *Federal Register* 20802) in the development of these ecological effects assessment methods. The purpose is to help ensure that ecological effects, in addition to health effects, will be considered in determining whether regulation is warranted.	This activity is ongoing. Under section 112(f), EPA is to consider the risk to public health remaining, or likely to remain, after sources are regulated under the section 112(d) MACT program. These additional standards, so-called "residual risk standards," will consider environmental as well as public health impacts. Under this effort, ecological effects assessment and screening methods are being developed.

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
k. EPA will evaluate whether other pollutants, including hexachlorobutadiene and methoxychlor, which are proposed Bioaccumulative Chemicals of Concern under the proposed Water Quality Guidance for the Great Lakes System (58 *Federal Register* 20802) and which have been identified as having potentially significant air sources, should be added to the list of Great Waters pollutants of concern.	This activity is ongoing. In addition to hexachlorobutadiene and methoxychlor, atrazine is also under consideration for addition to the list of Great Waters pollutants of concern.
l. EPA is continuing to emphasize pollution prevention as the goal in the development of control measures to reduce emissions of Great Waters pollutants of concern and is encouraging any voluntary pollution prevention and other emission reduction efforts.	There are a number of ongoing EPA activities that emphasize pollution prevention as a goal in the development of control measures. These activities are described in detail in Chapter IV of this report and include such projects as the Virtual Elimination Pilot Project, the Great Lakes Binational Toxics Strategy, development of Lakewide Management Plans (LAMPs), and the Great Lakes Water Quality Guidance.

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
m. In the development of regulations and pollution prevention or reduction strategies under the 1990 CAA Amendments, EPA will examine the potential for reductions of oxides of nitrogen and will determine how additional nitrogen oxide (NO_x) reductions can be achieved for protection of coastal water quality and related resources.	EPA's Integrated NO_x Strategy is described in the staff working draft document entitled "Nitrogen Oxides Impacts on Public Health and the Environment" (U.S. EPA 1997). This strategy will coordinate control efforts to maximize environmental benefits of reductions in ozone precursors, fine particulates, acidic deposition and eutrophication. The document was distributed to the CAA Advisory Committee at their December 5, 1996 meeting and public comments were requested by January 31, 1997. The document was also described at an Ozone Transport Assessment Group (OTAG) meeting and placed on OTAG's TTN web site. Several National Estuary Program sites are investigating the role of atmospheric deposition of nitrogen compounds in eutrophication. Another EPA activity called the Clean Air Power Initiative (CAPI) produced an October 1996 report that summarizes a strategy to cost-effectively reduce emissions of NO_x, SO_x, and mercury from utility boilers. CAPI information is available on the web site: www.epa.gov/capi. EPA has also proposed new ozone and particulate matters standards (December 13, 1996). EPA plans to complete the final rulemaking by July 19, 1997. These rules are expected to significantly reduce NO_x emissions, as will the revised NO_x New Source Performance Standard (NSPS) for utility and non-utility units. EPA is required to propose the NSPS by July 1, 1997.

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
n. EPA will develop Alternative Control Technology documents (ACTs) for NO_x. This is expected to result in nationwide NO_x emissions reductions, thus protecting coastal waters, as states develop regulations under the National Ambient Air Quality Standards program (NAAQS).	The ACT documents were required under section 183 of the CAA. These documents describe a wide range of NO_x control technologies for nine specific source categories (cement manufacturing, gas turbines, glass manufacturing, internal combustion engines, iron and steel, nitric/adipic acids, non-utility boilers, process heaters, and utility boilers). Great Waters funds were used to develop two of these documents. The purpose of the ACT documents is to help states adopt rules to meet the NO_x Reasonable Available Control Technology (RACT) requirements by May 31, 1995. In addition, the ACT documents should help states that develop beyond-RACT rules related to ozone attainment plans. Copies of the ACT documents are available from the National Technical Information Service at 1-800-553-NTIS.
2. EPA recognizes the need for an integrated multimedia approach to the problem of atmospheric deposition of pollutants to waterbodies and, therefore, will consider authorities beyond the Clean Air Act to reduce human and environmental exposure to Great Waters pollutants of concern.	
a. EPA will establish a funding and operational mechanism for all appropriate offices to pool their resources (both dollars and personnel) to more effectively and efficiently manage this multimedia problem. The Great Waters Core Project Management Group will serve as the liaison among EPA's Assistant Administrators (AAs) and Regional Administrators (RAs). Through this group, commitments will be obtained from each of the AAs and RAs to earmark funds for implementing the recommendations of the First Report to Congress or to take a lead role in the implementation of specific recommendations.	Evaluation of the opportunities for pooling resources between EPA offices showed that there was no available funding mechanism to do so, and that it would be impractical to develop such a mechanism. The Core Advisory Work Group agreed instead that the participating offices/agencies would integrate Core Group input into the planning for Great Waters-related efforts within their offices/agencies, as well as providing their input in planning Great Waters-funded activities. This integration of activities is ongoing.

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
b. EPA should use the discretionary authority in existing statutes to regulate or prohibit multimedia environmental releases that cause or contribute to a water quality impairment. The Administration wants to work with Congress (e.g., on Clean Water Act reauthorization) to develop approaches that would allow effective pollution control where other federal environmental statutes are not effective and where an integrated multimedia approach is the most efficient means to reduce unacceptable risk. This would not apply to mobile sources or pesticide programs. EPA would use the most appropriate existing environmental statute (e.g., the Clean Air Act for air releases) for controlling the release and would take into account the factors of revised section 307(a)(2) of the Clean Water Act.	In 1997, efforts to reauthorize the Clean Water Act are still ongoing.
c. Congress, with technical support from EPA, should develop legislation to prohibit the exportation of any pesticide product which contains an active ingredient that has been banned for all or virtually all uses in the United States. The recommendation to prohibit the export of banned pesticides was presented in the *Report of the National Performance Review: Creating a Government That Works Better and Costs Less*.	Such legislation was introduced into a Congressional committee in 1995, and failed to pass.

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
d. EPA will work with other countries to explore possible alternatives to reduce or eliminate the production, export, and use of pesticides banned in the United States.	This activity is ongoing. EPA currently participates in the North American Commission for Environmental Cooperation, which was set up as a parallel agreement to the North America Free Trade Agreement. In that group, EPA is a member of the Working Group on the Sound Management of Chemicals, which has established task forces on mercury, PCBs, DDT, and chlordane. EPA also represents the United States in negotiations on the Long-Range Transport of Air Pollution (LRTAP), which includes a number of these substances. In the near future, EPA will be representing the program on the United Nations Environmental Program negotiations on Persistent Organic Pollutants, the list for which is now under consideration, but which will likely include the LRTAP substances, as well as some others. EPA is currently participating in the negotiations for another United Nations treaty on Prior Informed Consent, which will control the international trade of many "delisted" (i.e., banned) substances.
e. EPA will explore the feasibility of creating an inventory of pesticide use within the United States and of establishing a program to identify and quantify stockpiles and emissions of pesticides of known and potential concern, including banned pesticides.	EPA believes that creating such an inventory is possible from annual information submitted under section 7 of the Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA, as amended by the Food Quality Protection Act of 1996). This information would cover basic producer and formulator inventory changes. Additional proprietary data sources could cover farm level inventories. While this type of inventory is feasible, EPA currently has no plans to compile this information.
f. EPA will continue to emphasize pollution prevention as a goal and to encourage voluntary pollution prevention efforts that lead to reductions in releases of Great Waters pollutants of concern. Several pollution prevention projects that address Great Waters pollutants of concern are currently underway:	The highlights of many of these efforts and milestones reached, including the PCB phaseout program and clean sweep actions, can be found in the document *Toward a Brighter Future, EPA Region 5, the First 25 Years, 1970-1995*, EPA-905-F-96-001, 1996.

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
– A "Virtual Elimination Pilot Project" is underway in the Great Lakes Basin, as part of a comprehensive toxics reduction effort. The Virtual Elimination (VE) Pilot Project proposes selecting a small group of toxics as a pilot and performing an in-depth analysis of opportunities for reduction from all sources.	The "pilot" portion of the VE project focused on the reduction of mercury and PCBs. A stakeholder meeting was held in the Great Lakes region in 1993. Based on the meeting, a draft report was developed by EPA to identify options to reduce mercury. A similar paper is currently being prepared by EPA to address PCBs. This project will continue with additional analyses of classes of substances rather than the use of a chemical-by-chemical approach.
– EPA has initiated a project to reduce risks from PCBs by asking all utilities in the Great Lakes area to voluntarily decommission their PCB electrical equipment.	Twelve major utilities in the Great Lakes basin conducted a study of the utility industry in EPA Region 5 and reported that the utilities have collectively removed almost 90 percent of the PCBs they had in service as of 1978. Individually, most of the 12 utilities indicated that they would continue efforts to remove PCB electrical equipment and several other utilities offered to assist with PCB phaseout outreach. In December 1995, Northern Indiana Public Service Company (NIPSCO) became the first utility to officially and formally commit to phase down its remaining PCB electrical equipment as part of the PCB Phasedown Program. Their commitment will involve the replacement or removal of all of their PCB equipment or PCB-contaminated oil in the equipment over the next 10 years with the vast majority of the PCBs being phased down within the next five years. As discussions on company-specific PCB reductions continue, EPA concurrently drafted a policy that could offer certain enforcement related credits to facilities that meet specific PCB phaseout targets. Once the policy is finalized, EPA expects renewed interest and participation in the PCB Phasedown Program.
– The Lake Superior Pollution Prevention Strategy was released in October 1993 as part of the Lake Superior Binational Program.	This effort has been completed.

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
– EPA, together with state Departments of Agriculture and local government agencies, has funded a series of "Clean Sweeps" to collect and properly dispose of existing stocks of canceled pesticides from residents in the Great Lakes area.	In the Lake Michigan basin, agricultural "clean sweeps" to properly collect and dispose of unused pesticides have been conducted in Indiana, Michigan, and Wisconsin. Also, a variety of pollution prevention and technical assistance projects have taken place in Milwaukee, Chicago, and western Michigan. EPA continues to fund agricultural clean sweeps on a modest but consistent level, encouraging states to develop innovative approaches to pesticide collections. Michigan, Minnesota, and Wisconsin are setting up permanent collection sites (similar to household hazardous waste sites) where pesticides will be collected over a number of years.
g. EPA will continue its work with Canada, under the Great Lakes Water Quality Agreement, on airborne toxic substances. These continuing bilateral efforts are assisting and will continue to assist in meeting Great Water program objectives during the 1990s.	EPA is continuing its work with Canada, as parties of the Great Lakes Water Quality Agreement, and with the activities of the International Joint Commission, as well as having Canadian input on major Great Waters planning and reporting activities. In April 1997, the United States and Canada also agreed to a strategic plan for eliminating toxic substances from the Great Lakes by 2006. The Great Lakes Binational Toxics Strategy calls for a number of milestones to be achieved from 1997 to 2006 including reductions in mercury, PCBs, and dioxins. The two countries have also agreed to boost bilateral cooperation to address pollution that crosses boundaries and to cooperate on environmental research and technology. A memorandum of understanding is to be signed in September 1997.
h. EPA will distribute technical information to state and local air and water agencies to facilitate cooperative efforts toward common goals to further reduce human and environmental exposure to Great Waters HAPs.	Outreach to state and local air and water agencies is an ongoing process. These efforts are exemplified by such programs as the Great Lakes Information Network, the Technology Transfer Network (TTN), the Regional Air Pollution Inventory Data System (RAPIDS), and other electronic information sources such as a number of EPA home pages on the World Wide Web. In addition, EPA continues to develop emission factors for state and local use in developing emission inventories. These documents are available electronically on the TTN.

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
i. EPA will initiate discussions about possible mechanisms that regional EPA offices and state agencies could use for sharing information on new or renewal permit applications for sources with the potential to emit Great Waters pollutants of concern.	This activity has yet to be initiated. If, in the course of new permit applications and renewals, there appear to be common issues among the states with respect to Great Waters pollutants, the regional EPA offices may initiate some mechanism to provide information exchange and consistency between the various permitting agencies. To date, with the early implementation of the permitting programs, this has not been an issue.

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
3. EPA will continue to support research activities and will develop and implement a strategy describing necessary research and policy assessments to address the mandates of section 112(m).	
a. EPA is developing a strategy to target research necessary to answer the scientific questions outlined in section 112(m). The strategy will be reviewed by EPA's Science Advisory Board and will influence decisionmaking on the priority and funding for future research. This strategy will focus on utilization of the mass balance approach for determining relative loading and will acknowledge the need for a balance between monitoring, modeling, and emission inventory efforts for that work. The strategy will also consider how to better identify those persistent chemicals with the tendency to bioaccumulate that may become problematic if emissions continue. Included in the strategy will be an assessment of the need for development of tools that can be used to: (1) assess and quantify the human health and environmental risk from exposure to air toxics, especially via indirect exposure routes, and (2) quantify the social, environmental, and economic benefits and costs of pollution prevention and regulatory actions.	EPA is working on a research strategy for the Great Waters program that focuses on mass balance work, modeling and monitoring support, control technologies and strategies, and development of assessment methods that can be used to evaluate waterbodies other than the Great Lakes. The purpose of such a strategy is to avoid duplication of effort in funded research and to target specific areas where research is needed to respond to the mandate of the Great Waters program. In addition, EPA's Mercury Task Force is in the process of developing an Agency research strategy specific to mercury. This effort will be fully coordinated with the Great Waters program.
b. EPA will continue to work with NOAA to pursue the development and application of the appropriate technical tools to further define and estimate loadings to the Great Waters and to identify sources of atmospherically deposited pollutants.	The Great Waters program continues to fund NOAA's development of transport and deposition models, as well as work to parameterize important atmospheric processes for those models. These efforts are exemplified by the development and application of ammonia and organic nitrogen measurement methods which will aid in understanding the effects, transport, and sources of various species of nitrogen. NOAA also assisted in the development of EPA's long-range transport analyses for mercury.
c. Through the use of Great Waters program funds and other resources, EPA will continue to support those research activities identified as priorities by the research communities and affirmed by the Great Waters Core Project Management Group.	Funding of appropriate research activities is an ongoing effort. As mentioned above, EPA will continue to identify and evaluate research priorities during development of a Great Waters program research strategy.

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
– EPA will continue work on the characterization of processes and parameters for mass balance modeling and the verification of the mass balance methodology, especially the development of the prototype mass balance program being conducted in Lake Michigan.	EPA has developed and funded, in cooperation with a number of other agencies and organizations, the Lake Michigan Mass Balance Study (LMMB). The LMMB is intended to develop the predictive capability to determine the environmental benefits of specific load reduction scenarios and the time needed to realize those benefits. For this study, the atmospheric deposition of toxics is being monitored and the concentrations of toxics in fish, phytoplankton, sediment, tributaries are being measured. Pollutants chosen for the LMMB are total mercury, atrazine, trans-nonachlor, and PCBs.
– EPA will work with state agencies to complete regional emission inventories for the Great Lakes and will complete a national screening level emission inventory for section 112(c)(6) chemicals (seven of the Great Waters pollutants), and will identify categories of sources of these pollutants.	Continuation of the Great Lakes regional emission inventory work is made possible by EPA CAA section 105 grants to the Great Lakes states, and for the section 112(c)(6) project by Great Waters contract funding. Four states completed a pilot study of major urban areas along the shore of Lake Michigan in December 1995 using RAPIDS. Work is continuing by all of the Great Lakes states to now build a comprehensive regional air toxics inventory for 49 air pollutants. Mobile source emissions will be added to the emissions inventory in the future.–EPA's inventory for section 112(c)(6) of the CAA was made publicly available for comment in October 1996. The final 112(c)(6) inventory and listing decisions will be completed by December 1997. The draft inventories can be obtained from EPA's Internet website at: www.epa.gov/oar/oaqps/airtox/112c6fac.html.
– EPA will continue source characterization and identification activities.	Source characterization work is ongoing in several areas, ranging from regulatory work by EPA under CAA section 112 to inventory development by state agencies. Other specific projects to characterize and identify emission sources include the Mercury Stack Testing Project in the Lake Superior Basin which will provide speciated mercury data for a number of different source types. Also, the Lake Michigan Urban Air Toxics Study (LMUATS) and the Atmospheric Exchange Over Lakes and Oceans Study (AEOLOS) focused on the southern Lake Michigan area to quantify and characterize wet and dry depositional fluxes of trace metals, PCBs, and PAHs resulting from emission sources in this urban area.

TABLE A-1
Status of Actions Recommended in First Report to Congress

Recommended Action	Status
– EPA will complete and evaluate mercury screening level deposition models using screening emission inventories and will determine whether to transfer the method to other chemicals and to provide support for other more intensive regional air emission inventory efforts.	EPA has performed and documented extensive mercury deposition modeling in the draft Mercury Study Report to Congress. Work is ongoing, as mentioned above, in transferring this methodology to other chemicals, notably POM, dioxin, and cadmium, on a regional scale using the REMSAD model. During 1997, these efforts will focus on multi-pollutant modeling on a regional scale.
– EPA will continue to support ongoing monitoring efforts.	EPA continues to support the Integrated Atmospheric Deposition Network (IADN). The network began collecting data in 1990. Currently, there are three master monitoring stations in the United States and two master monitoring stations in Canada. Other satellite sites have also been added in both countries. Data collection has proceeded at all sites, and research and development of sampling and analytical methodologies is ongoing. A workshop to discuss the results of the IADN work to date will be held in June 1997.
d. EPA will initiate discussions among the appropriate groups to identify ongoing benefits analysis efforts and human health (cancer and noncancer) and environmental risk assessment efforts within the Agency, in other federal programs, in other countries, in academia, and elsewhere. The goal is to define more clearly the research/data needs and to develop a long-term plan for developing tools and methods for benefits analyses and risk assessments.	This activity will be initiated in 1997.

TABLE A-2
Emission Standards Addressing Great Waters Pollutants of Concern

Standard	Pollutants Controlled
Asphalt Hot-Mix Production	Polycyclic organic matter (POM)
Asphalt Roofing Production	POM
Battery Production	Mercury
Carbamate Insecticides Production	POM
Carbon Reactivation Furnaces	Dioxin
Carbon Black Production	Mercury, POM
Chlorine Production	Mercury
Chlorinated Solvents Production	Hexachlorobenzene
Coke Ovens: Charging, Topside & Door Leaks	POM
Coke Ovens: Pushing, Quenching & Battery Stacks	POM
Commercial Coal Combustion	Mercury, POM
Commercial Natural Gas Combustion	POM
Commercial Oil Combustion	Mercury, POM
Commercial Wood/Wood Residue Combustion	POM
Crematories	Mercury, POM
Dental Preparation and Use	Mercury
Drum and Barrel Reclamation	Dioxins, POM
Electrical Apparatus Manufacturing	Mercury
Ferroalloy Manufacture	POM
Fluorescent Lamp Recycling	Mercury
Gasoline Distribution (Aviation)	Lead
Gasoline Distribution (Stage I)	Lead, POM
Gasoline Distribution (Stage II)	Lead, POM
General Laboratory Activities	Mercury
Geothermal Power	Mercury
Hazardous Waste Incineration	Dioxins, mercury, PCBs, POM
Industrial Coal Combustion	Mercury, POM
Industrial Natural Gas Combustion	POM
Industrial Oil Combustion	Mercury, PCBs, POM
Industrial Stationary IC Engines - Diesel	POM

TABLE A-2
Emission Standards Addressing Great Waters Pollutants of Concern

Standard	Pollutants Controlled
Industrial Stationary IC Engines - Natural Gas	POM
Industrial Waste Oil Combustion	POM
Industrial Wood/Wood Residue Combustion	Dioxins, POM
Instrument Manufacturing	Mercury
Iron and Steel Foundries	Dioxins, POM
Lamp Breakage	Mercury
Landfill (Gas) Flares	Dioxins, mercury
Lightweight Aggregate Kilns	Dioxins, mercury
Lime Manufacturing	Mercury
Medical Waste Incineration	Cadmium, dioxins, mercury, PCBs, POM
Municipal Waste Combustion	Cadmium, dioxins, mercury, PCBs, POM
Naphthalene - Miscellaneous Uses	POM
Naphthalene Production	POM
Naphthalene Sulfonates Production	POM
Non-Residential Wood Combustion	Mercury
Non-Road Vehicles and Equipment (NRVE) - Aircraft	POM
NRVE - Other	Lead, POM
On-Road Vehicles	Dioxins, POM
Open Burning of Scrap Tires	POM
Other Biological Incineration	Dioxin, PCBs, POM
Pesticides Application	Hexachlorobenzene
Pesticides Manufacture	Hexachlorobenzene
Petroleum Refining-Catalytic Cracking Units	POM
Phthalic Anhydride Production	POM
Portland Cement Manufacture: Hazardous Waste Kilns	Dioxins, mercury, POM
Portland Cement Manufacture: Non-Hazardous Waste Kilns	Dioxins, mercury, POM
Primary Aluminum Production	POM
Primary Copper Production	Cadmium, mercury
Primary Lead Smelting	Mercury
Pulp and Paper - Kraft Recovery Furnaces	Dioxins, POM

TABLE A-2
Emission Standards Addressing Great Waters Pollutants of Concern

Standard	Pollutants Controlled
Pulp and Paper - Lime Kilns	POM
Pulp and Paper - Sulfite Recovery Furnaces	POM
Residential Coal Combustion	Dioxins, mercury, POM
Residential Natural Gas Combustion	POM
Residential Oil Combustion	Dioxins, mercury, POM
Residential Wood Combustion	Dioxins, POM
Scrap or Waste Tire Incineration	Dioxins, PCBs, POM
Secondary Aluminum Smelting	Dioxins
Secondary Copper Smelting	Dioxins
Secondary Lead Smelting	Dioxins, POM
Secondary Mercury Production	Mercury
Sewage Sludge Incineration	Dioxins, mercury, PCBs, POM
Stationary Gas Turbines - Diesel	POM
Stationary Turbines - Natural Gas	POM
Utility Coal Combustion	Dioxins, mercury, POM
Utility Natural Gas Combustion	POM
Utility Oil Combustion	Dioxins, mercury, PCBs, POM
Wildfires and Prescribed Burning	Dioxins, POM
Wood Treatment/Wood Preserving	Cadmium, dioxins, POM

APPENDIX B
FISH CONSUMPTION ADVISORIES

As summarized in Chapter II of this report, fish advisory data for each state are collected in a national data base. The information in the data base available for use in this report was current through 1995 (U.S. EPA 1996b). For each advisory, the data base contains information such as waterbody name, waterbody type (e.g., estuary, Great Lake, river), pollutant name, fish species, population targeted by the advisory (called advisory type in this report), advisory status (e.g., active), and a state contact name and telephone number. The data base does not, however, contain information on the levels of pollutants in fish or the benchmark levels set by a particular state for each advisory type.

For this report, EPA reviewed the data base for any active fish advisories related to the 15 Great Waters pollutants of concern in the Great Lakes, Lake Champlain, Chesapeake Bay, and several coastal waters.[1] (The data base does not include in its list of pollutants three of the Great Waters pollutants of concern: α-HCH, nitrogen, and TCDF (furans).) Other criteria used to select fish advisory data for this report include:

- This report includes advisory data only for those Great Waters pollutants of concern listed by name in the data base. For example, "metals" is a pollutant included in the data base, but this report includes advisories associated only with specific listings for the metals "cadmium," "lead," and "mercury."

- For the Great Lakes, only advisories for which the waterbody type was designated in the data base as "Great Lake" are included. For example, the waterbody type for an entire Great Lake (e.g., Lake Huron) and for portions of the lake (e.g., Saginaw Bay) are designated as "Great Lake," as are the major Great Lakes connecting channels (e.g., Detroit River). Note, however, that the tables of fish consumption advisories in Section II.A of this report contain only lakewide advisories. Table B-1, below, includes the advisories for tributaries or portions of the waterbody that were designated as "Great Lake," as well as lakewide advisories.

- The advisories for coastal waters represent the advisories as they are identified in the data base. Therefore, advisories for some coastal waters represent the entire waterbody (e.g., Tampa Bay), while others represent smaller estuaries or portions of the waterbody (e.g., Baltimore Harbor). Table B-1 provides details on the waterbodies that represent the coastal water advisories. The coastal waters for which advisory data are presented in this report represent only a small sample of coastal waters for which there are fish consumption advisories.

- This report does not include statewide advisories. Florida, Massachusetts, Maine, New Jersey, New York, and Rhode Island all have issued statewide advisories, which are reported in the data base, that may affect the Great Waters (e.g., Narragansett Bay in Rhode Island).

[1] The data base contains fish advisory data for numerous other pollutants and several pollutant mixtures (e.g., metals, organic compounds, pesticides) other than the Great Waters pollutants of concern. In addition, all U.S. waterbodies in which there are fish advisories are included in the data base, not only the Great Waters.

Section II.A of this report discusses the five main types of advisories included in the fish advisory data base. Data on informational health advisories, no-kill zones, and commercial fishing bans are summarized in Chapter II and are not repeated in this appendix. This appendix provides detailed information on the advisories to limit fish consumption and those to restrict fish consumption.

This detailed fish consumption advisory information is summarized in Table B-1 for the Great Lakes, Lake Champlain, Chesapeake Bay, and other selected coastal waters. This information was used to develop the fish consumption advisory tables in Section II.A of this report. For each waterbody, Table B-1 presents the state issuing the advisory, the relevant Great Waters pollutants of concern, the fish species affected by the advisory, and the advisory type. The two types of fish consumption advisories (limit fish consumption and restrict fish consumption) are issued for two different populations (the general population and subpopulations potentially at risk), resulting in four categories of fish consumption advisories:

- Advisories to subpopulations potentially at risk to restrict the size and frequency of meals of particular species (RSP);

- Advisories to the general population to restrict the size and frequency of meals of particular species (RGP);

- Advisories to subpopulations potentially at risk (e.g., pregnant or nursing women, small children) to not consume particular species (NCSP); and

- Advisories to the general population to not consume particular species (NCGP).

For the most recent fish advisory data for waterbodies in a particular state or for more details on how the state sets its advisories, the state office responsible for coordinating the fish advisory program in that state should be contacted.

TABLE B-1
Fish Consumption Advisories in the Great Waters

Waterbody[a]	Great Waters Pollutant of Concern	Fish Species	Advisory Type[b]
GREAT LAKES			
Lake Erie			
MI	PCBs	Carp, catfish	NCGP
OH	PCBs	Carp, channel catfish, chinook salmon > 19", coho salmon, freshwater drum, lake trout, smallmouth bass, steelhead trout, walleye, white bass, white perch	RGP
PA	Chlordane, PCBs	Carp, channel catfish, lake trout	NCGP
Maumee Bay (OH)	PCBs	Channel catfish Carp	NCGP RGP

TABLE B-1
Fish Consumption Advisories in the Great Waters (continued)

Waterbody[a]	Great Waters Pollutant of Concern	Fish Species	Advisory Type[b]
Lake Huron			
MI	Chlordane, dioxins	Lake trout > 22" Lake trout < 22"	NCGP NCSP, RGP
	PCBs	Lake trout > 22" Brown trout > 21", lake trout	NCGP NCSP, RGP
Saginaw Bay (MI)	Chlordane	Lake trout > 22"	NCGP
	Dioxins	Lake trout > 22"	NCGP, NCSP, RGP
	PCBs	Carp, channel catfish, lake trout > 22" Brown trout > 21", lake trout > 22"	NCGP NCSP, RGP
Lake Michigan			
IL	Chlordane, PCBs	Brown trout > 23", chinook salmon > 32", lake trout > 23" Brown trout < 23", chinook salmon 21-32", coho salmon > 26", lake trout 20-23"	NCGP NCSP, RGP
IN	PCBs	Brown trout > 23", carp, catfish, chinook salmon > 32", lake trout > 23" Brown trout < 23", chinook salmon 21-32", coho salmon > 26", lake trout 20-23"	NCGP NCSP, RGP
MI	Chlordane	Lake trout > 23", lake whitefish > 23" Lake trout 20-25"	NCGP NCSP, RGP
	Mercury	Walleye > 22"	RGP, RSP
	PCBs	Brown trout > 23", carp, channel catfish Lake trout > 23"	NCGP NCSP, RGP
WI	Chlordane	Lake trout > 23" Lake trout 20-23"	NCGP NCSP, RGP
	PCBs	Brown trout > 23", carp, catfish, chinook salmon > 32", lake trout > 23" Brown trout < 23", chinook salmon 21-32", coho salmon > 26", lake trout 20-23"	NCGP NCSP, RGP
Green Bay (MI)	PCBs	Brook trout > 15", brown trout > 21", carp, northern pike > 28", rainbow trout > 22", splake trout > 20", sturgeon, walleye > 20", white bass Brown trout < 21", splake trout < 20"	NCGP NCSP, RGP
Green Bay (WI)	PCBs	Brook trout > 15", brown trout > 12", carp, chinook salmon > 25", northern pike > 28", rainbow trout > 22", splake trout > 16", walleye > 20", white bass Splake trout < 16"	NCGP NCSP, RGP
Little Bay de Ngoc (MI)	Mercury	Walleye > 22"	RGP, RSP
	PCBs	Brown trout > 23", carp, channel catfish Lake trout > 23", longnose sucker > 16"	NCGP NCSP, RGP

TABLE B-1
Fish Consumption Advisories in the Great Waters (continued)

Waterbody[a]	Great Waters Pollutant of Concern	Fish Species	Advisory Type[b]
Lake Michigan (continued)			
Old North Harbor, Waukegan (IL)	Chlordane, PCBs	Alewife, carp	NCGP
Lake Ontario			
NY	Dioxins, PCBs	American eel, carp, channel catfish, chinook salmon, lake trout, white perch Brown trout > 20", coho salmon > 21", rainbow trout > 25" Brown trout, coho salmon, rainbow trout, white sucker Brown trout < 20", coho salmon < 21", rainbow trout < 25", white perch, white sucker	NCGP, NCSP NCGP NCSP RGP
Irondequoit Bay (NY)	PCBs	Carp	NCGP, NCSP
Lake Superior			
MI	Chlordane, mercury, toxaphene	Ciscowet > 18"	NCGP
	PCBs	Ciscowet > 18", lake trout > 30" Lake trout 20-30"	NCGP NCSP, RGP
MN	PCBs	Ciscowet > 20" Brown trout, chinook salmon, coho salmon, ciscowet < 20", lake herring, lake trout, lake whitefish, rainbow trout	NCGP RGP
WI	Chlordane	Ciscowet > 20"	NCGP
	Mercury	Walleye 26-30" Walleye 18-26" Walleye < 18"	NCGP NCSP, RGP RSP
	PCBs	Ciscowet > 20", lake trout > 30"	NCGP
Chequamegon Waters (WI)	Mercury	Largemouth bass 15-18" Northern pike > 30" Largemouth bass < 15", northern pike < 30"	NCGP NCSP, RGP RSP
Thunder Bay (MI)	Chlordane, dioxins	Lake trout > 22" Lake trout < 22"	NCGP NCSP, RGP
	PCBs	Lake trout > 22" Brown trout > 21", carp, lake trout < 22"	NCGP NCSP, RGP

TABLE B-1
Fish Consumption Advisories in the Great Waters (continued)

Waterbody[a]	Great Waters Pollutant of Concern	Fish Species	Advisory Type[b]
Connecting Channels			
Detroit River (MI)	Mercury	Freshwater drum > 14"	RGP, RSP
	PCBs	Carp	NCGP
Lake St. Clair (MI)	Mercury	Muskellunge, sturgeon Bluegill sunfish > 8", brown bullhead catfish > 14", carpsucker > 18", freshwater drum > 14", largemouth bass, northern pike > 26", rock bass > 8", smallmouth bass > 18", walleye > 20", white bass > 13", white perch > 10"	NCGP RGP, RSP
	PCBs	Channel catfish > 22" Carp > 22"	NCGP NCSP, RGP
Niagara River (NY)	Dioxins, PCBs	American eel, carp, channel catfish, chinook salmon, lake trout, white perch Brown trout > 20", coho salmon > 21", rainbow trout > 25" Brown trout, coho salmon, rainbow trout, smallmouth bass, white sucker Brown trout < 20", coho salmon < 21", rainbow trout < 25", smallmouth bass, white sucker	NCGP, NCSP NCGP NCSP RGP
	PCBs	Carp	RGP
St. Clair River (MI)	Mercury	Freshwater drum > 14"	RGP, RSP
	PCBs	Carp > 21", gizzard shad > 10"	NCSP, RGP
St. Lawrence River (NY)	Dioxins, PCBs	American eel, carp, channel catfish, chinook salmon, lake trout Brown trout > 20", coho salmon > 21", rainbow trout > 25" Brown trout, coho salmon, rainbow trout White perch Brown trout < 20", coho salmon < 21", rainbow trout < 25"	NCGP, NCSP NCGP NCSP NCSP, RGP RGP
	PCBs	Fish	NCGP, NCSP
St. Mary's River (MI)	Mercury	Walleye > 19"	RGP, RSP
LAKE CHAMPLAIN			
NY	Mercury	Walleye > 19"	NCSP, RGP
	PCBs	Brown bullhead, eel, lake trout > 25", yellow perch	NCSP, RGP
VT	Mercury	Walleye	NCSP, RGP
	PCBs	Lake trout > 25"	NCSP, RGP

TABLE B-1
Fish Consumption Advisories in the Great Waters (continued)

Waterbody[a]	Great Waters Pollutant of Concern	Fish Species	Advisory Type[b]
SELECTED COASTAL WATERS			
Chesapeake Bay			
Anacostia River/ Potomac River (DC)	Chlordane, PCBs	American eel, carp, channel catfish	RGP
Back River (MD)	Chlordane	American eel, channel catfish	RGP
Baltimore Harbor (MD)			
Lake Roland, Jones Falls (MD)	Chlordane	Black crappie, carp	RGP
Delaware Bay			
Delaware Estuary (DE)	PCBs	Channel catfish, striped bass, white catfish, white perch Channel catfish, striped bass, white catfish	NCGP RGP, RSP
Delaware River and Delaware Estuary (PA)	Chlordane, PCBs	American eel, channel catfish, white perch	NCGP
Galveston Bay			
Houston Ship Channel (TX)	Dioxins	Blue crab, catfish	NCSP, RGP
Long Island Sound			
CT	PCBs	Bluefish > 25", striped bass	NCSP, RGP
NY	Chlordane	Striped bass	RGP
	PCBs	Striped bass	NCGP
NY/NJ Harbor			
Arthur Kill (NJ), Kill van Kull (NJ)	Dioxins	Blue crab, striped bass	NCGP, NCSP
	PCBs	American eel, bluefish > 24" > 6 lbs, striped bass, white catfish, white perch	NCSP, RGP
Newark Bay (NJ)	Dioxins	Blue crab, striped bass Bluefish > 24" > 6 lbs, striped bass	NCGP, NCSP RGP
	PCBs	White catfish, white perch Bluefish > 24" > 6 lbs, striped bass	NCSP, RGP NCSP
Raritan Bay (NJ), Raritan River (NJ), Sandy Hook Bay (NJ)	Dioxins	Blue crab	RGP, RSP
	PCBs	American eel, bluefish > 24" > 6 lbs, striped bass, white catfish, white perch	NCSP, RGP
Tidal Passaic River (NJ)	Dioxins	Crustaceans, fish, shellfish	NCGP, NCSP
	PCBs	Striped bass American eel, bluefish > 23" > 6 lbs, white catfish, white perch	NCGP, NCSP NCSP, RGP

TABLE B-1
Fish Consumption Advisories in the Great Waters (continued)

Waterbody[a]	Great Waters Pollutant of Concern	Fish Species	Advisory Type[b]
San Francisco Bay			
San Francisco Bay delta region (CA)	Dioxins, DDT, mercury, PCBs	Striped bass > 35" Sharks > 24", striped bass > 27" Fish except anchovy, herring, salmon, and smelt	NCGP NCSP RGP, RSP
Tampa Bay (FL)	Mercury	Crevalle jack, gafftop sail catfish, ladyfish, spanish mackerel	RGP, RSP

[a] For the Great Lakes, the advisories are listed first by lakewide advisories, in alphabetical order by state, followed by portions of the waterbody and major tributaries that were designated in the data base as "Great Lake." Lake Champlain advisories are all lakewide. For coastal waters, including Chesapeake Bay, the advisories are presented by how the coastal water was designated in the data base, which may be by full name or by waterbody portion.

[b] NCGP = advises against consumption by the general population
NCSP = advises against consumption by subpopulations potentially at risk (e.g., pregnant or nursing women, small children)
RGP = advises the general population to restrict size and frequency of meals of the particular species
RSP = advises subpopulations potentially at risk to restrict size and frequency of meals of the particular species

Source: U.S. EPA 1996b.